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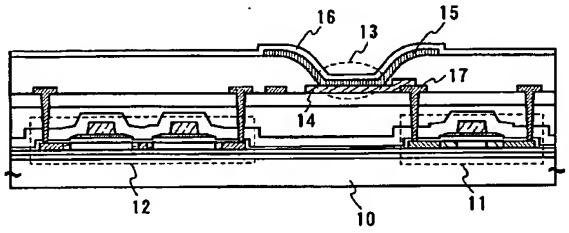
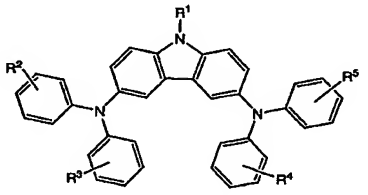
(54) Title: CARBAZOLE DERIVATIVE, LIGHT EMITTING ELEMENT, AND LIGHT EMITTING DEVICE





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(57) Abstract: It is an object of the present invention to provide a carbazole derivative that has a great carrier transporting property and a great light emitting property, permits an uniform film to be formed, and that is unlikely to undergo crystallization and morphologically stable. In addition, it is an object of the present invention to provide a light-emitting element from which stable light emission can be obtained efficiently for a long stretch of time by using the carbazole derivative. A carbazole derivative represented by the general formula (1) is synthesized. By applying this material to alight-emitting element, it becomes possible to provide a light-emitting element from which stable light emission can be obtained efficiently for a long stretch of time. [General Formula (1) is inserted.]

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For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

DESCRIPTION

CARBAZOLE DERIVATIVE, LIGHT EMITTING ELEMENT, AND LIGHT EMITTING DEVICE

TECHINICAL FIELD

The present invention relates to a carbazole derivative, and further relates to a light-emitting element that has an anode, a cathode, and a layer including an organic compound from which luminescence can be obtained by applying an electric field (hereinafter, referred to as "a layer including a luminescent layer").

BACKGROUND ART

Organic compounds include more varied material kinds of materials in comparison with inorganic compounds, and have a possibility that a material that has various functions can be synthesized by an appropriate molecular design. Also, they have features that a molded article such as a film is flexible and excellent workability is provided by polymerization. Based on these advantages, photonics and electronics utilizing functional organic materials have been attracting attention recently.

For example, examples of a photoelectric device utilizing an organic semiconductor material as a functional organic material include a solar cell and a light-emitting device (also referred to as an organic electroluminescent device), which are devices utilizing an electric property (carrier transporting property) and an optical property (light absorption or light emission) of the organic semiconductor material, and, among them, the light-emitting device has been showing remarkable progresses.

The light-emitting device has a light-emitting element interposing a layer including a luminescent material between a pair of electrodes (an anode and a cathode), which is said to have the light emission mechanism that a hole injected from the anode

and an electron injected from the cathode are recombined in the luminescence center the layer including the luminescent material to form an excited state when a voltage is applied between the both electrodes and energy is released to emit light while moving back from the excited state toward the ground state. As the excited state, a singlet excited state and a triplet excited state are known, and luminescence is said to be possible through any of the singlet excited state and the triplet excited state.

The layer including the luminescent material can have a single layer structure of only a light-emitting layer including a luminescent material; However, the layer including the luminescent material can be formed by laminating not only a light-emitting layer but also layers such as a hole injecting layer, a hole transporting layer, a hole blocking layer, an electron transporting layer, and an electron injecting layer. As for the light-emitting layer, by doping a host material with a guest material, it is possible to appropriately change a color of luminescence. In addition, depending on the combination of a host material and a guest material, there is a possibility of improving the luminance and life of luminescence.

As materials to be used for the layer including the luminescent material, a lot of materials that have various structures and functions are used, which includes a material including a carbazole moiety that has great photoconductivity (carbazole derivatives), for example. Specifically, carbazole derivatives such as CBP (4, 4 - di (N - carbazole) biphenyl) and PVK (polyvinylcarbazole) are known, which are often used today.

CBP that is a low molecular weight material is deposited mainly by evaporation, and is often used as a host material that has a hole transporting property in a light-emitting layer (for example, refer to Patent Document 1).

(Patent Document 1) Japanese Patent Laid-Open No. 2001-244077

However, while having a feature of a high thermal property value (great heat resistance), this material has disadvantages that it is hard to keep a film amorphous and

the film is likely to undergo crystallization when this material is used to form the film.

On the other hand, PVK that is a high molecular weight material is deposited mainly by a wet method such as coating (including spin coating) or inkjet, and is often used as a host material in a light-emitting layer in the same way as CBP (for example, refer to Patent Document 2).

(Patent Document 2) Japanese Patent Laid-Open No. 2001-257076

The high molecular weight material has disadvantages of poor heat resistance and low reliability in addition to a disadvantage that the method for deposition is limited while having a property of being superior to the low molecular weight material in luminance characteristic (the highest luminance of several tens of thousands cd/m^2) in the case of using a host material in a light-emitting layer.

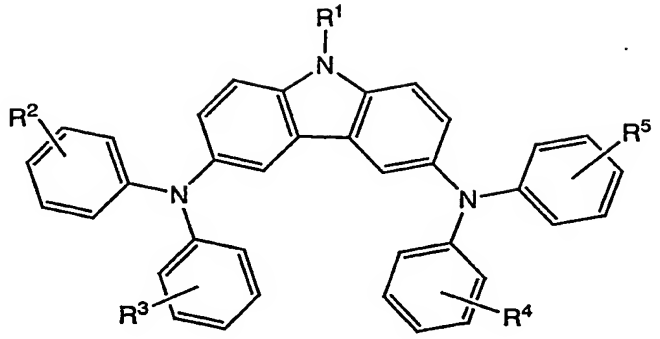
Accordingly, since each of the low molecular weight material and the high molecular weight material, including a carbazole moiety, has the disadvantages described above, material development for overcoming these disadvantages is desired in order to further improve characteristics of a light-emitting element.

DISCLOSURE OF INVENTION

It is an object of the present invention to provide a carbazole derivative that has a great carrier transporting property and a great light emitting property, permits an uniform film to be formed, and that is unlikely to undergo crystallization and morphologically stable. In addition, it is an object of the present invention to provide a light-emitting element from which stable light emission can be obtained for a long stretch of time and a light-emitting device using the light-emitting element.

A lot of earnest studies of the inventors have finally found out that a carbazole derivative represented by the following general formula (1) has a great carrier transporting property and a great light emitting property and is unlikely to crystallize. Accordingly, an aspect of the present invention is a carbazole derivative represented by

the following general formula (1).



(1)

(where R_1 is one of hydrogen, halogen, a cyano group, an alkyl group having 1 to 20 carbon atoms, a haloalkyl group having 1 to 20 carbon atoms, an alkoxyl group having 1 to 20 carbon atoms, a substituted or unsubstituted aryl group, and a substituted or unsubstituted heterocyclic group, and R_2 to R_5 may be identical or different, and are individually one of hydrogen, halogen, a cyano group, an alkyl group having 1 to 20 carbon atoms, an alkoxyl group having 1 to 20 carbon atoms, an acyl group having 1 to 20 carbon atoms, a haloalkyl group having 1 to 20 carbon atoms, a dialkylamino group having 1 to 20 carbon atoms, a diarylamino group having 1 to 20 carbon atoms, a substituted or unsubstituted heterocyclic group, and a carbazolyl group.)

By using the carbazole derivative mentioned above, a light-emitting element from which stable light emission can be obtained efficiently for a long stretch of time can be provided. Accordingly, another aspect of the present invention is a light-emitting element including the carbazole derivative mentioned above. Since the carbazole derivative according to the present invention has a great hole transporting property, a light-emitting element including the carbazole derivative mentioned above as a hole transporting material is preferable.

Further, the carbazole derivative according to the present invention has a feature of showing a light-emitting property. Therefore, a light-emitting element using the carbazole derivative according to the present invention as a luminescent material

can be formed.

Further, the carbazole derivative according to the present invention has a feature of a large energy gap. Therefore, the carbazole derivative can be used as a host material to form a light-emitting layer in combination with a guest material.

In the case mentioned above, the carbazole derivative according to the present invention is preferable particularly in the case of using a phosphorescent material requiring a host material with a large energy gap as a guest material.

By implementing the present invention, a carbazole derivative that has a great carrier transporting property and a great light emitting property, permits an uniform film to be formed, and is unlikely to undergo crystallization and morphologically stable, can be obtained. In addition, by using the carbazole derivative, it is possible to obtain a light-emitting element from which stable light emission can be obtained efficiently for a long stretch of time.

BRIEF DESCRIPTION OF DRAWINGS

In the accompanying drawings:

Fig. 1 is a diagram illustrating a light-emitting device;

Fig. 2 is a diagram showing a fluorescence spectrum of a carbazole derivative;

Fig. 3 is a diagram showing a UV-Vis absorption spectrum of the carbazole derivative;

Fig. 4 is a diagram showing a ^{1}H NMR spectrum of a carbazole derivative;

Fig. 5 is an enlarged view of the ^{1}H NMR spectrum of the carbazole derivative;

Fig. 6 is a diagram showing a fluorescence spectrum of the carbazole derivative;

Fig. 7 is a diagram showing a UV-Vis absorption spectrum of the carbazole derivative;

Fig. 8 is a diagram showing a ^{1}H NMR spectrum of a carbazole derivative;

Fig. 9 is a diagram showing a ^{13}C NMR spectrum of the carbazole derivative;

Fig. 10 is an enlarged view of the ^{13}C NMR spectrum of the carbazole derivative;

Fig. 11 is a diagram a diagram showing a fluorescence spectrum of the carbazole derivative;

Fig. 12 is a diagram showing a UV-Vis absorption spectrum of the carbazole derivative;

Fig. 13 is a diagram illustrating a structure of a light-emitting element according to the present invention;

Fig. 14 is a diagram illustrating a structure of a light-emitting element according to the present invention;

Figs. 15A and 15B are diagrams illustrating a light-emitting device.

Figs. 16A to 16C are diagrams illustrating electronic devices.

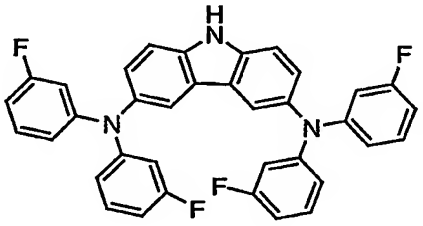
BEST MODE FOR CARRYING OUT THE INVENTION

[Embodiment 1]

A carbazole derivative according to the present invention has a structure represented by the above-mentioned general formula (1). Specific examples of R_1 include hydrogen, halogen elements such as fluorine and chlorine, cyano groups, alkyl groups such as a methyl group, an ethyl group, an isopropyl group, and a cyclohexyl group, haloalkyl groups such as a trifluoromethyl group, alkoxyl groups such as a methoxy group, an ethoxy group, an isopropoxy group, and a cyclohexyloxy group, aryl groups such as a phenyl group, a naphthyl group, and an anthryl group, and heterocyclic groups such as an imidazolyl group, an oxathiolyl group, and a thiazolyl group. R_2 to R_5 may be identical or different, and specific examples of R_2 to R_5 include hydrogen, halogen elements such as fluorine and chlorine, cyano groups, alkyl groups such as a methyl group, an ethyl group, an isopropyl group, and a cyclohexyl group, alkoxyl

groups such as a methoxy group, an ethoxy group, an isopropxy group, and a cyclohexyloxy group, acyl groups such as an acetyl group, an acroyl group, a malonyl group, a benzoyl group, and a naphthoyl group, haloalkyl groups such as a trifluoromethyl group, dialkylamino groups such as a dimethylamino group, a diethylamino group, and a diisopropyl amino group, diarylamino groups such as a diphenylamino group, heterocyclic groups such as an imidazolyl group, an oxathiolyl group, and a thiazolyl group, and carbazolyl groups.

In addition, as specific examples of carbazole derivatives to be formed according to the present invention, by appropriately changing the structures of R_1 to R_5 in the general formula (1), for example, carbazole derivatives represented by the following structure formulas (2) to (75) can be formed by appropriately changing the structures of R_1 to R_5 in the general formula (1). However, the present invention is not to be considered limited to these.



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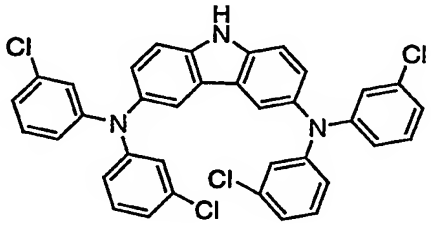

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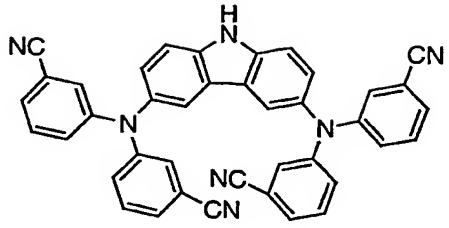

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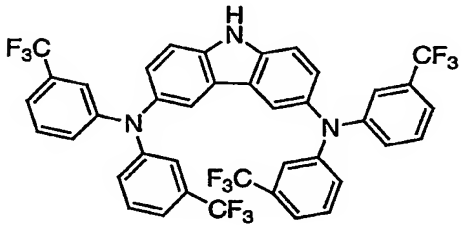

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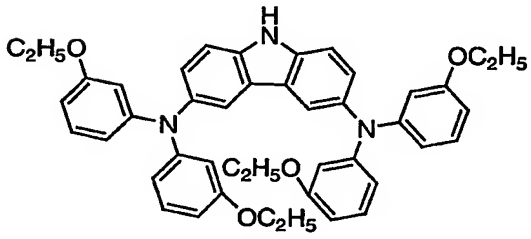

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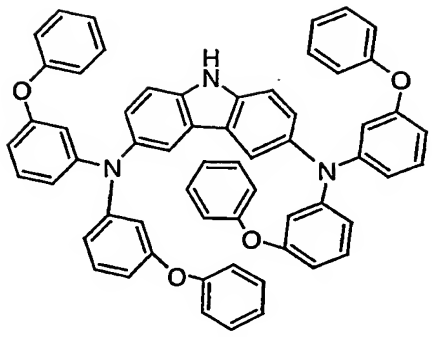

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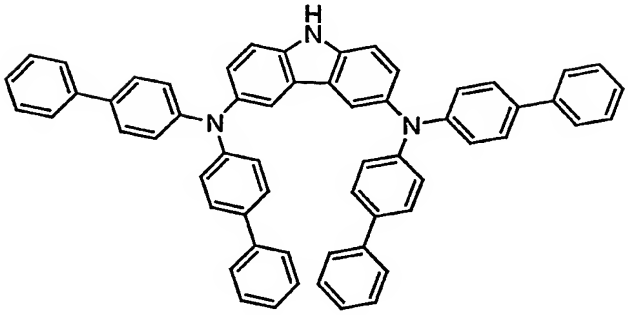

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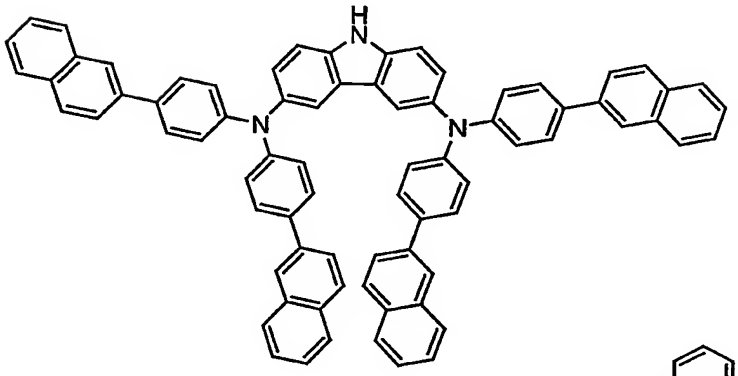

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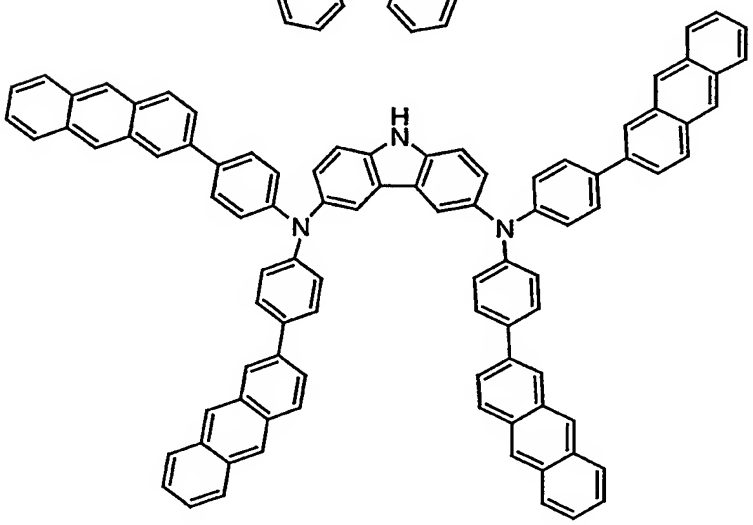

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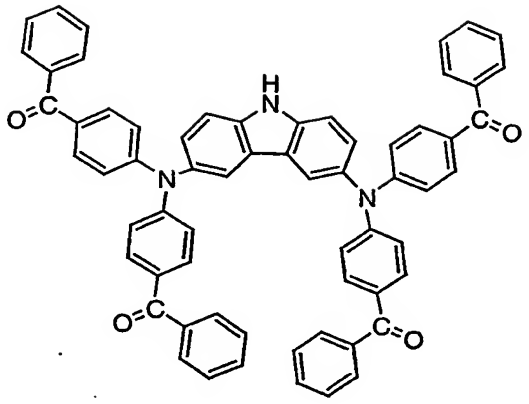

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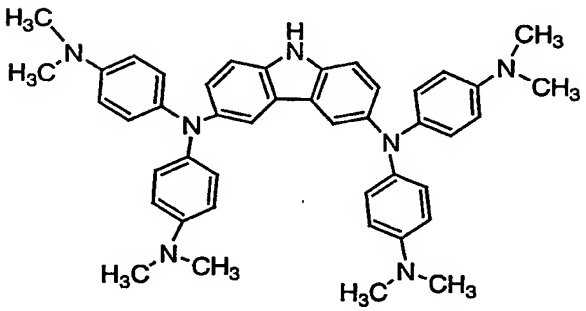

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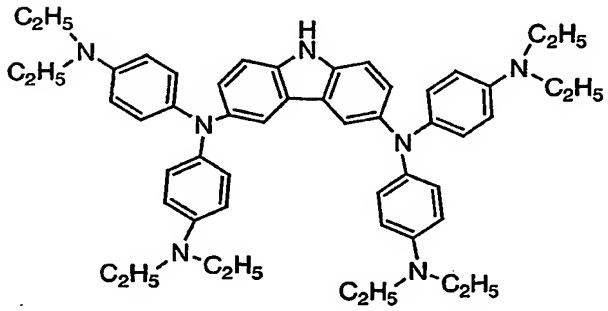

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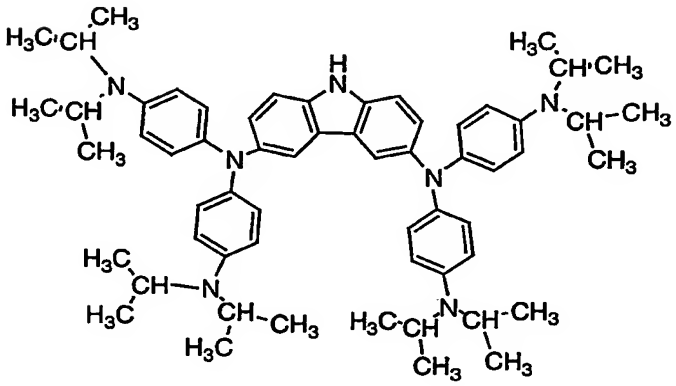

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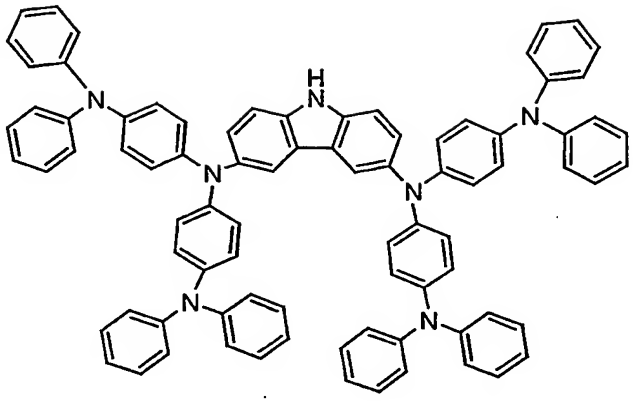

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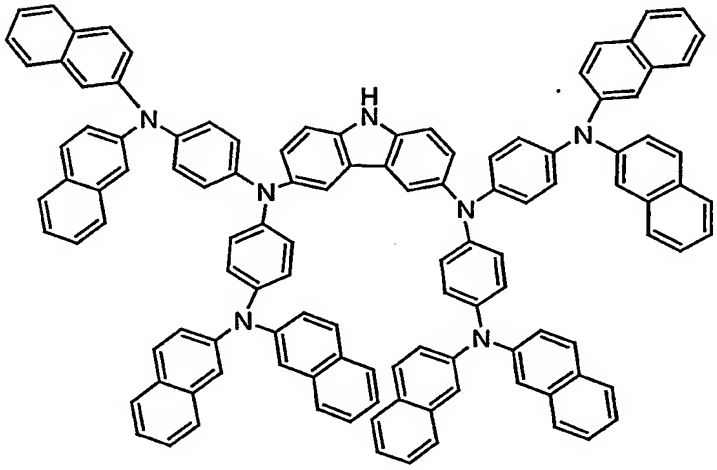

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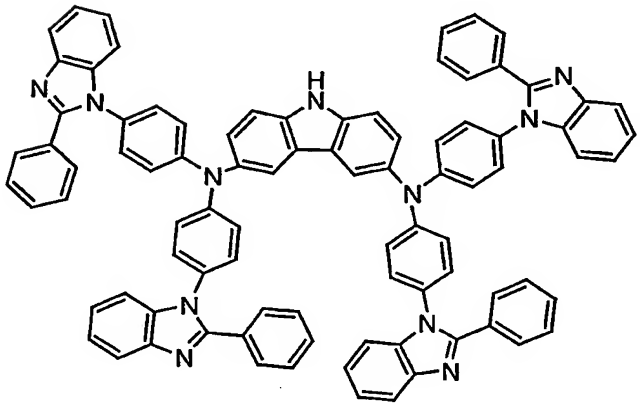

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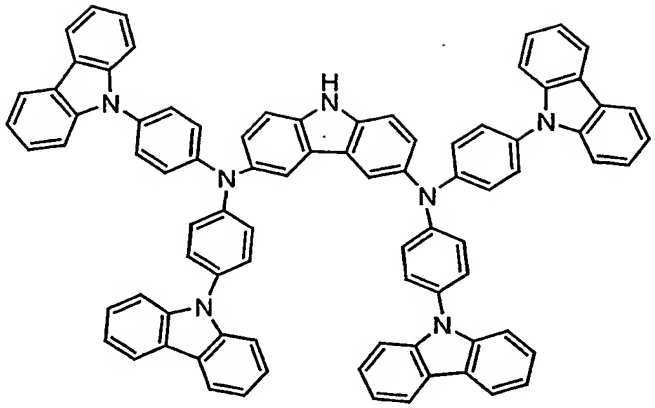

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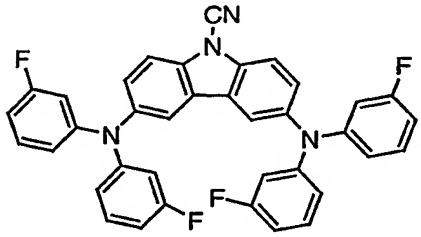

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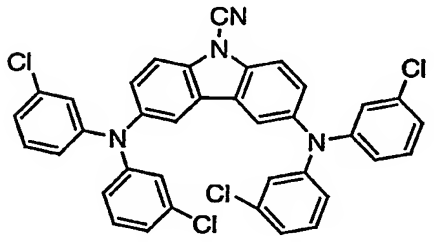

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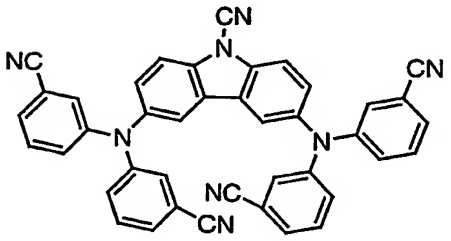

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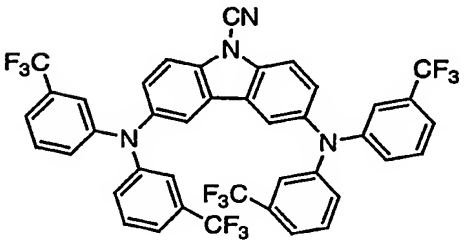

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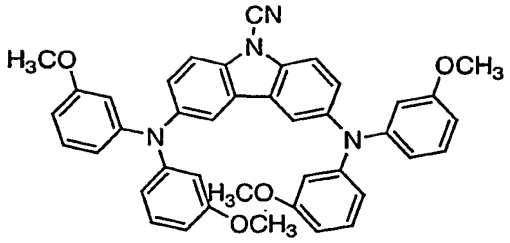

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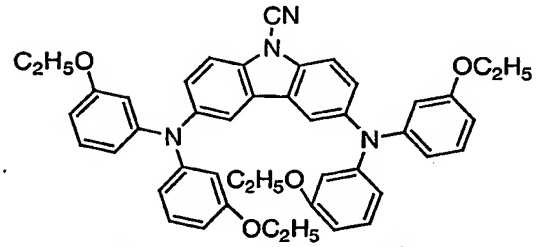

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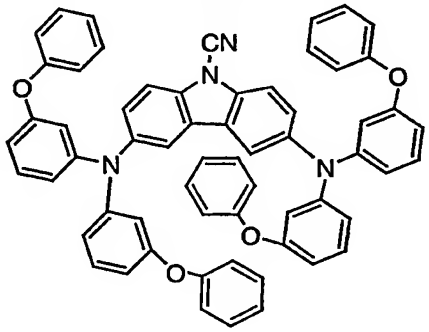

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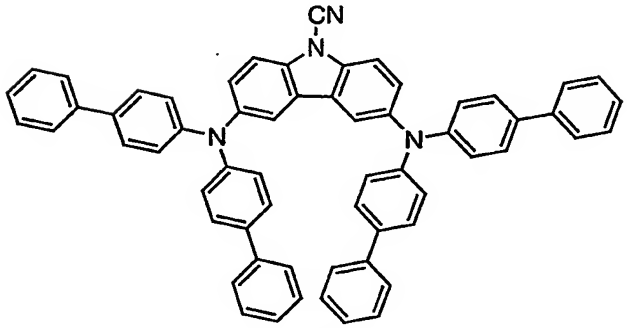

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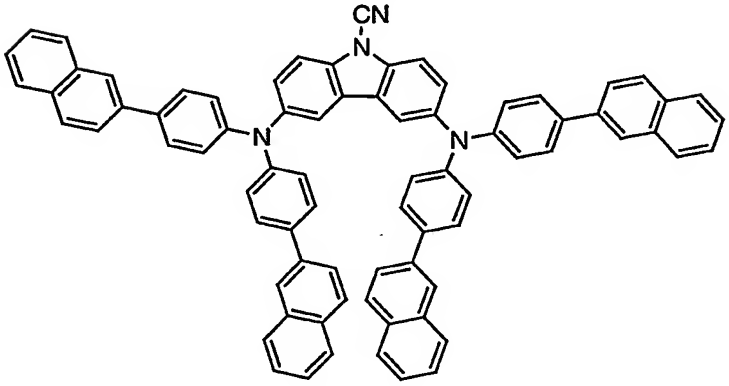

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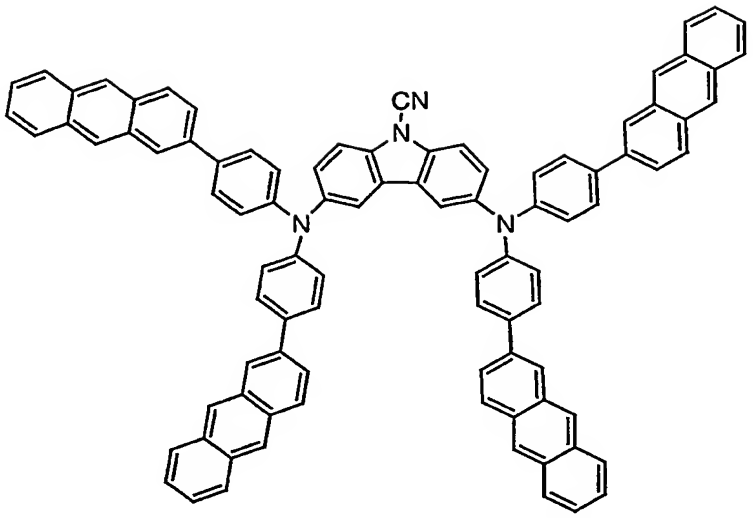

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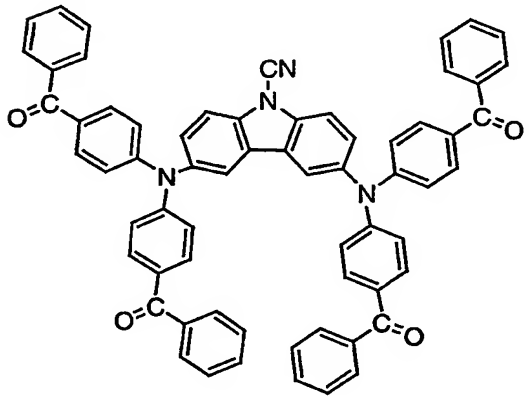

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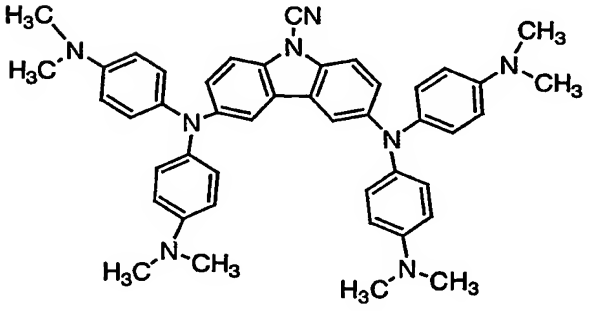

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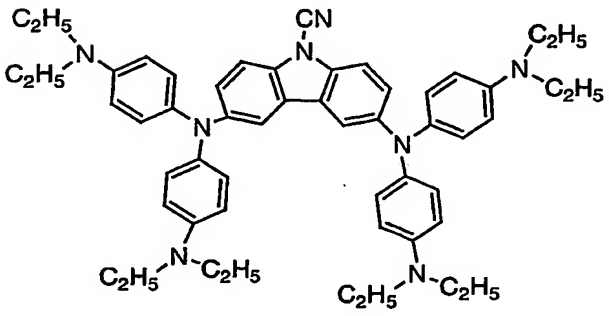

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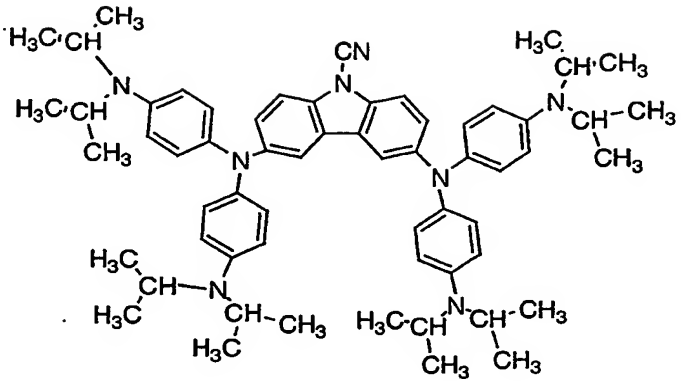

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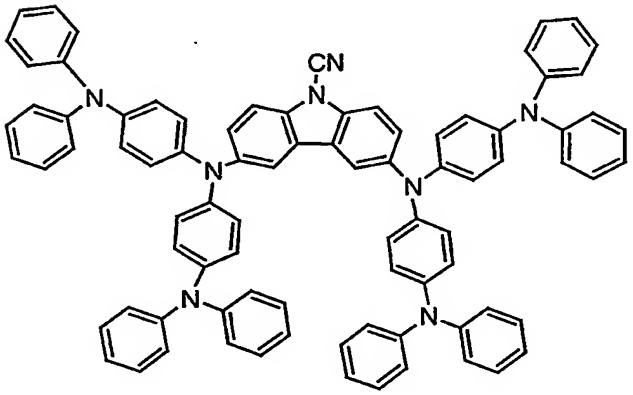

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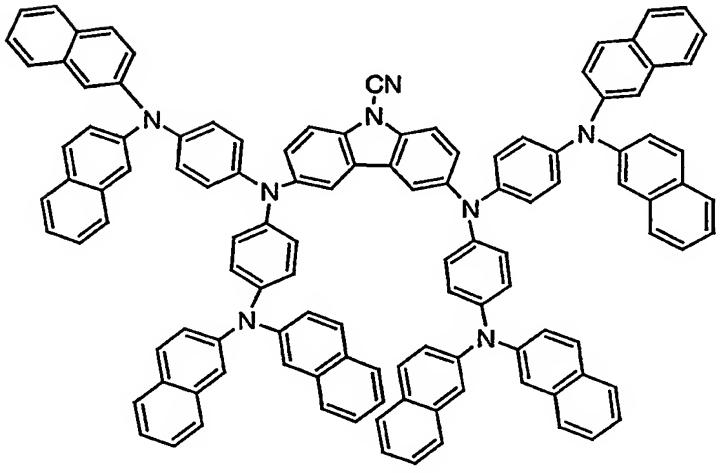

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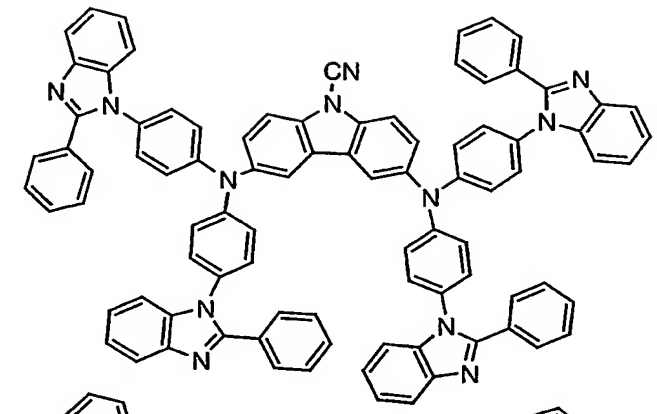

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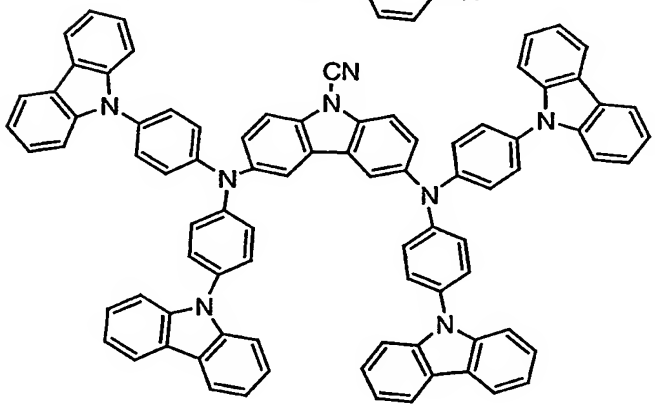

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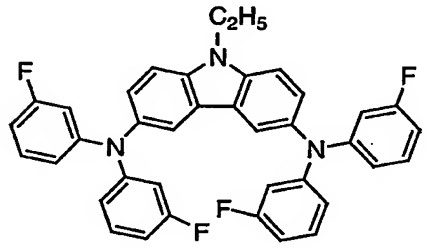

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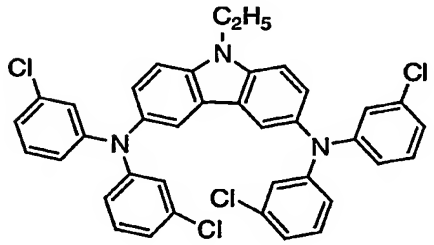

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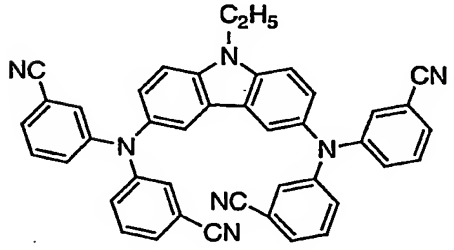

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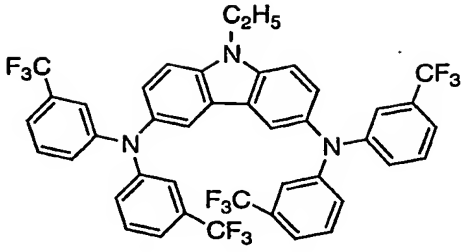

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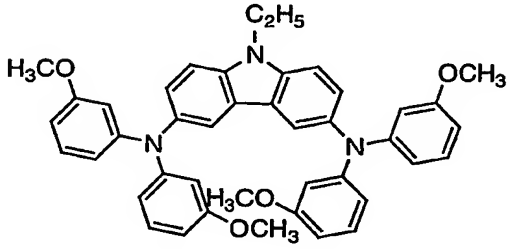

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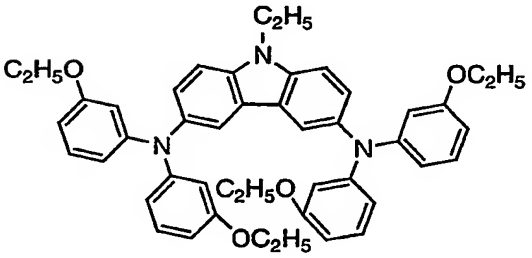

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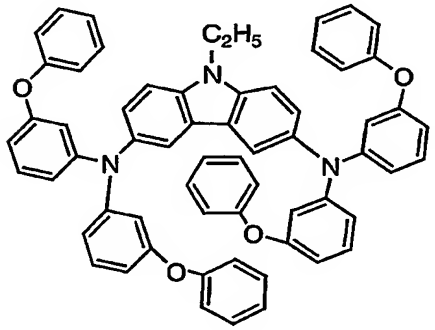

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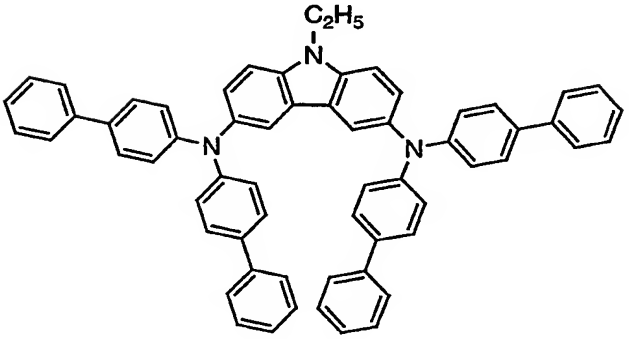

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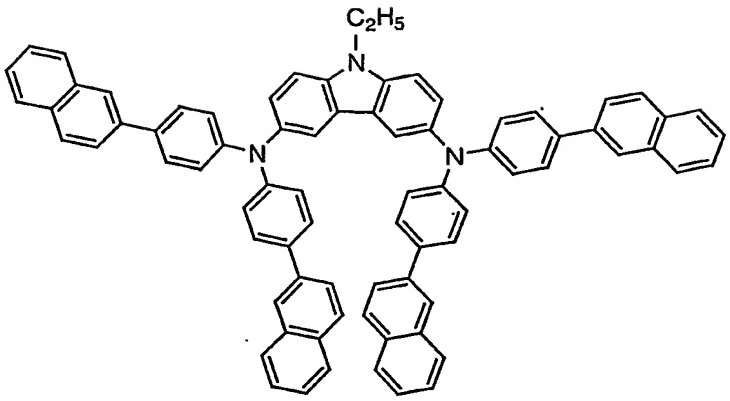

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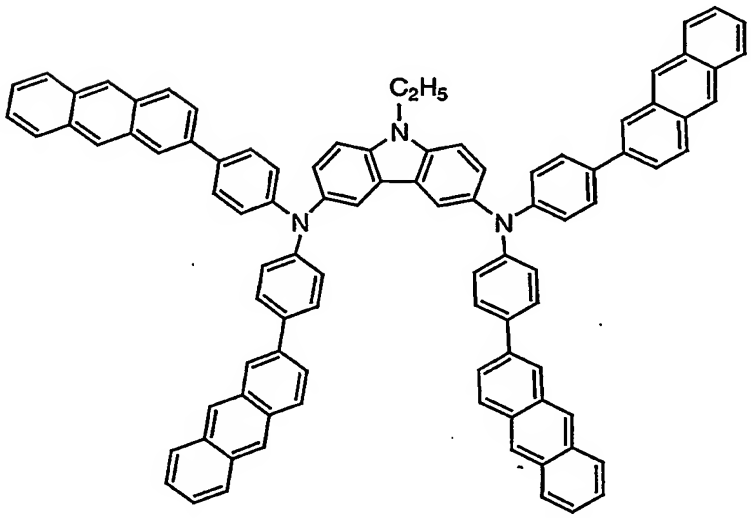

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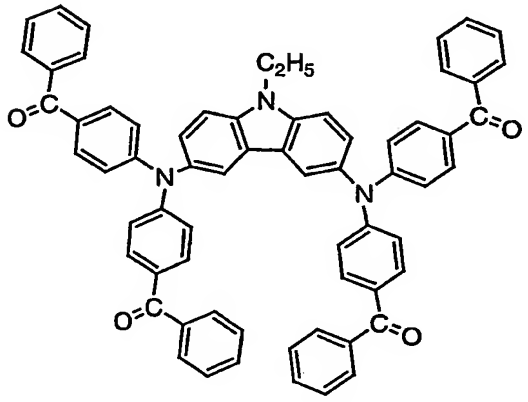

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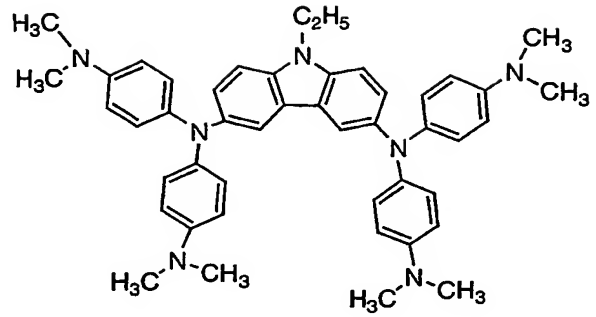

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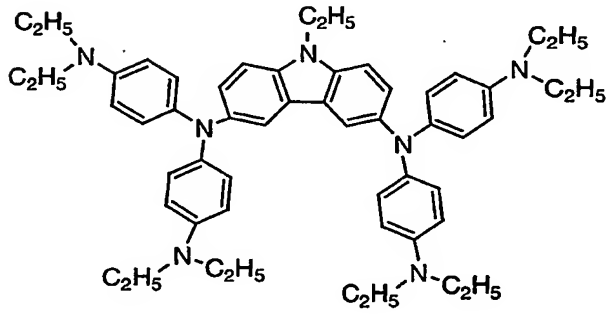

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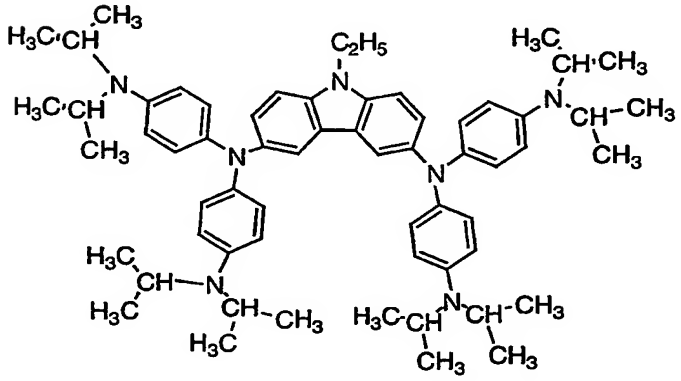

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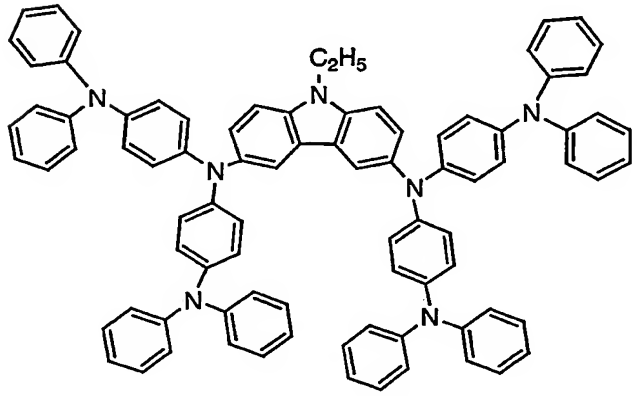

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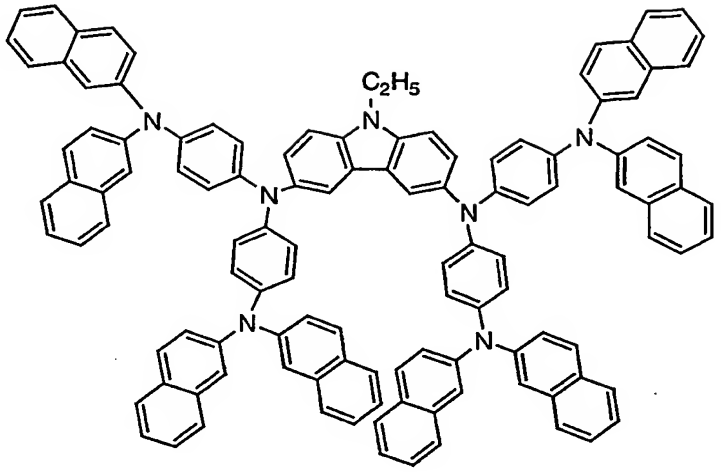

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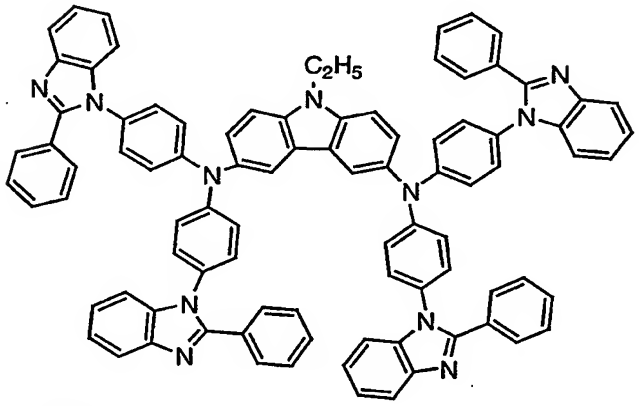

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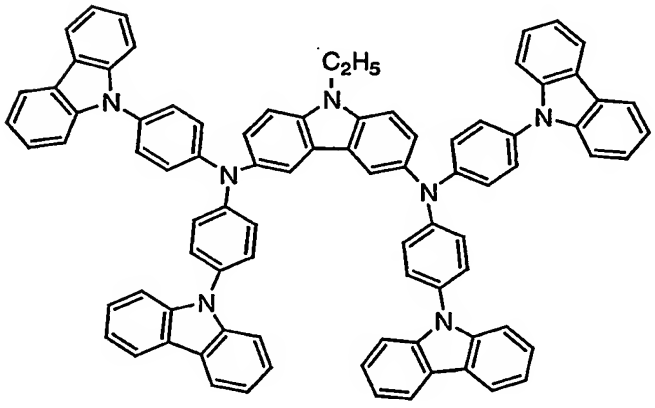

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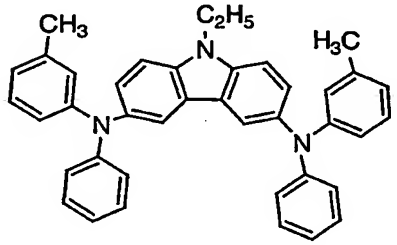

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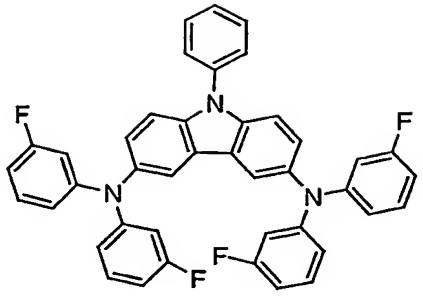

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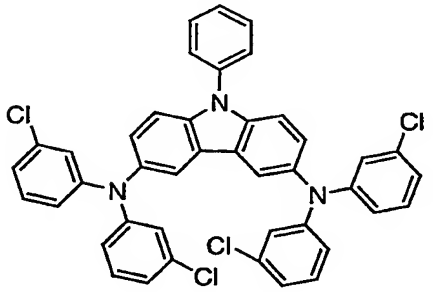

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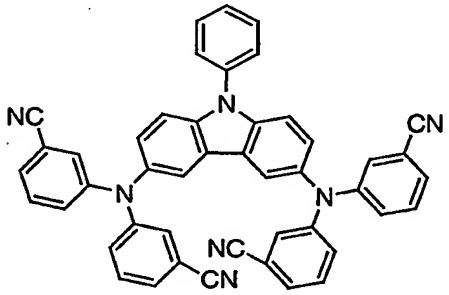

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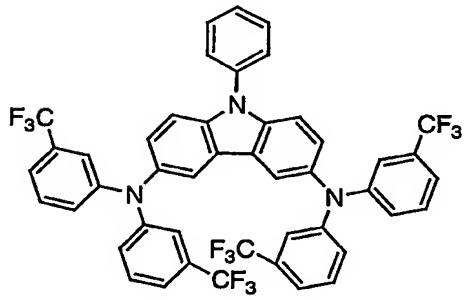

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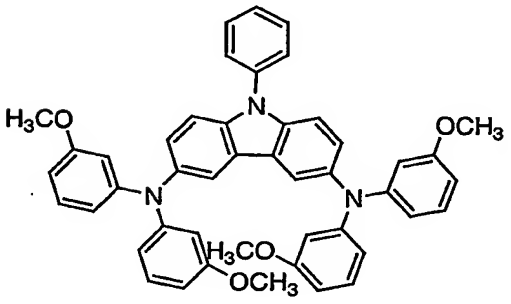

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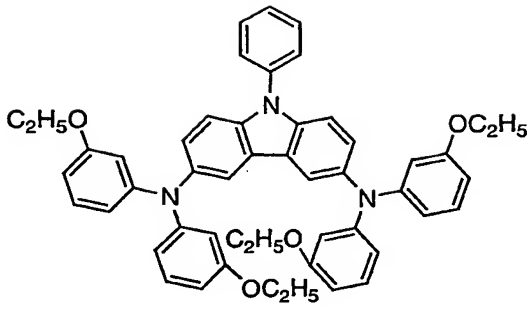

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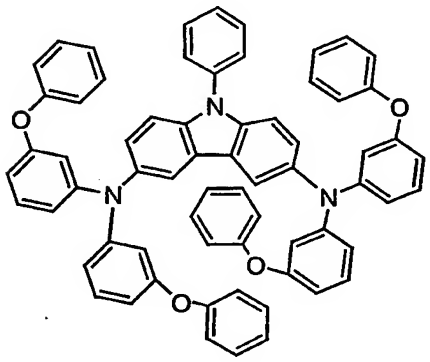

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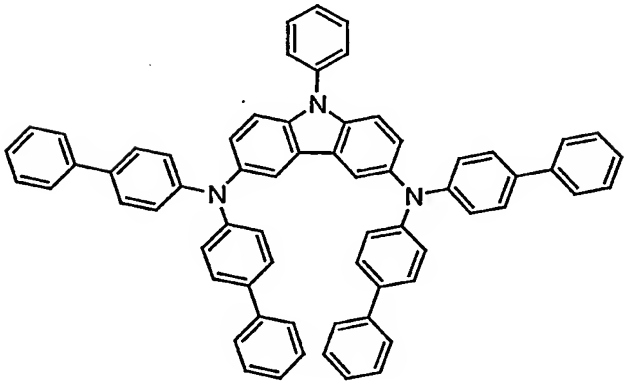

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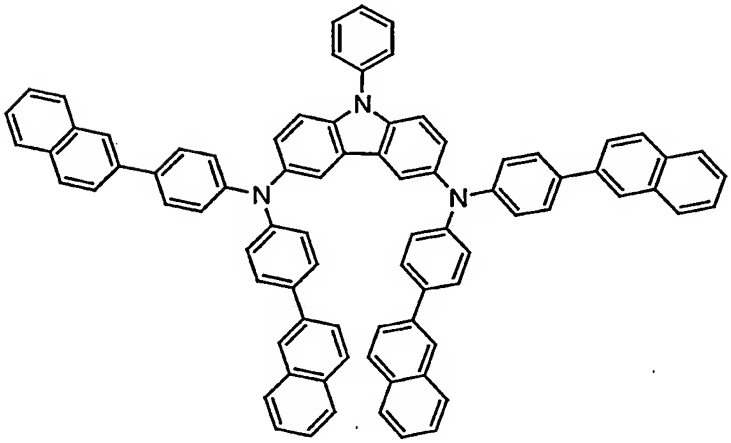

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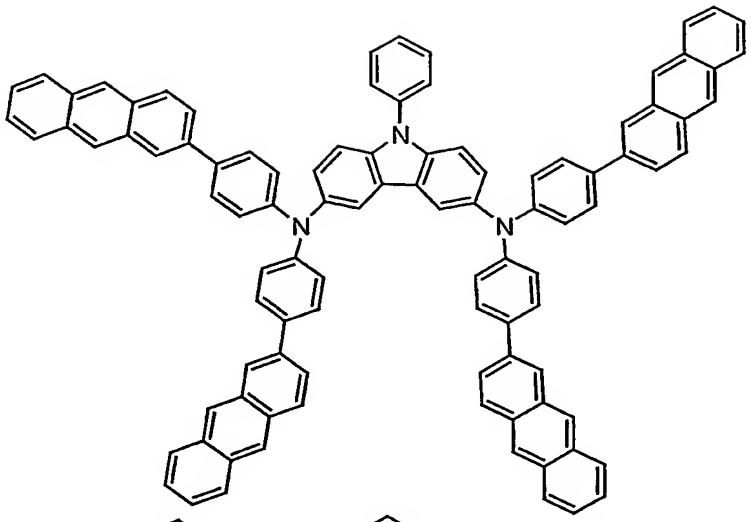

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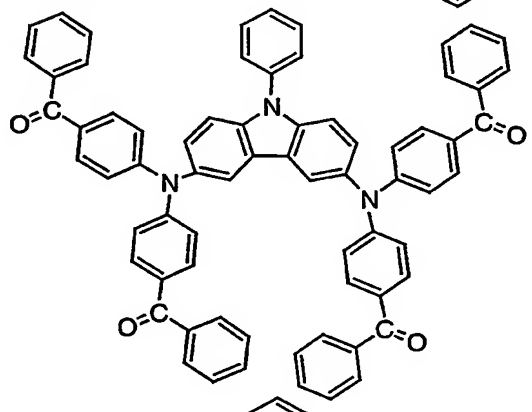

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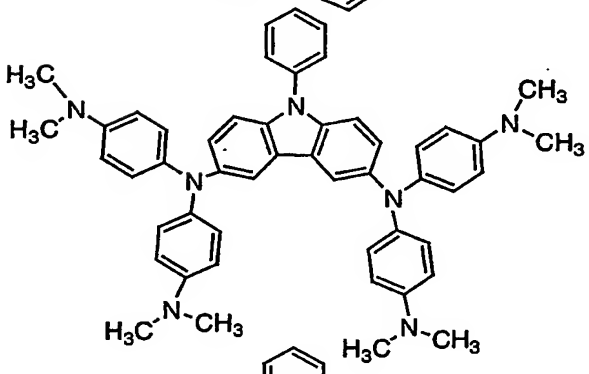

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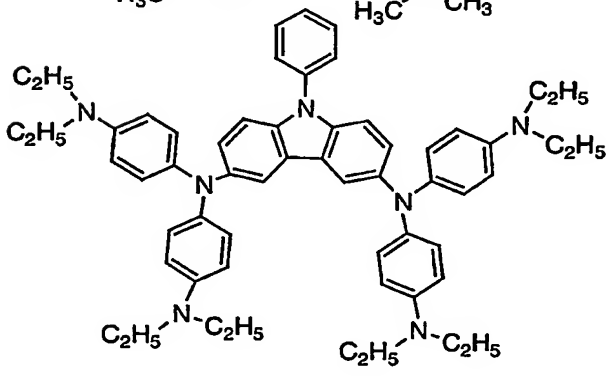

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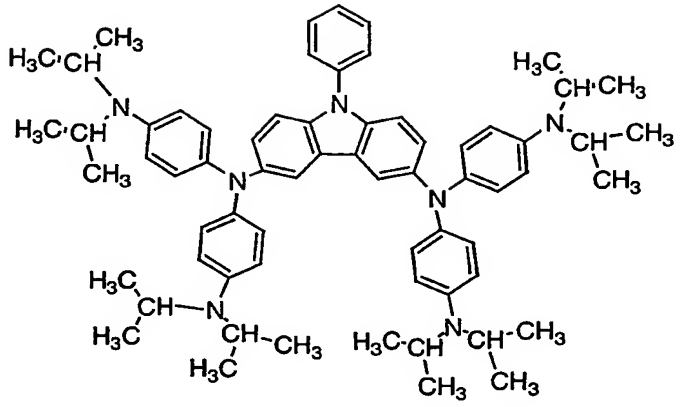

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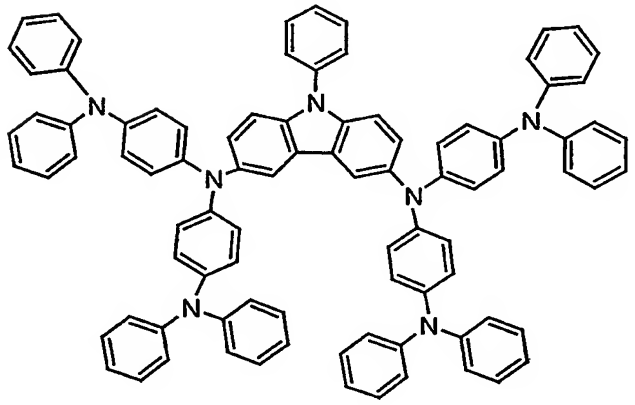

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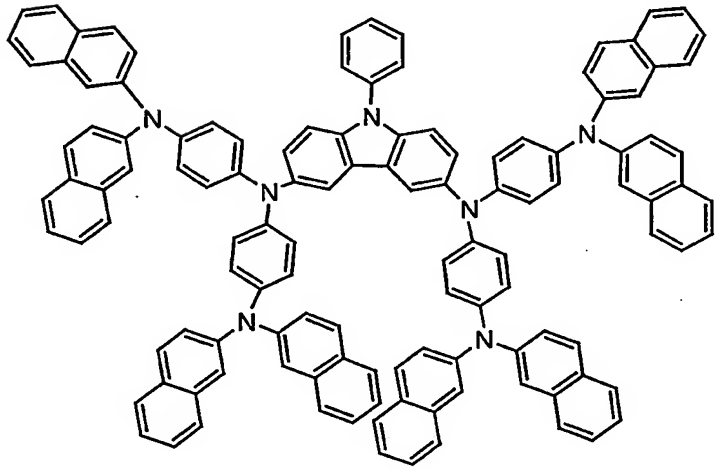

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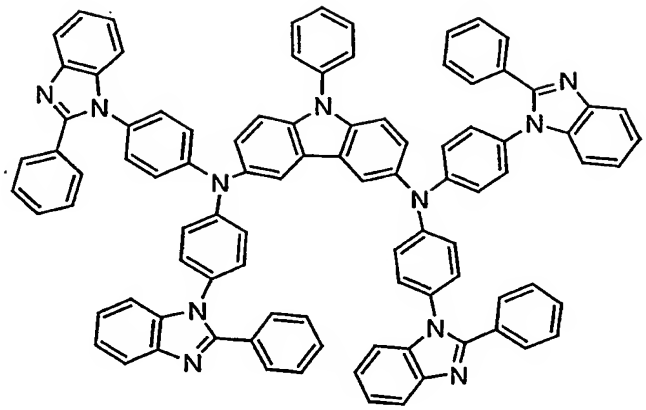

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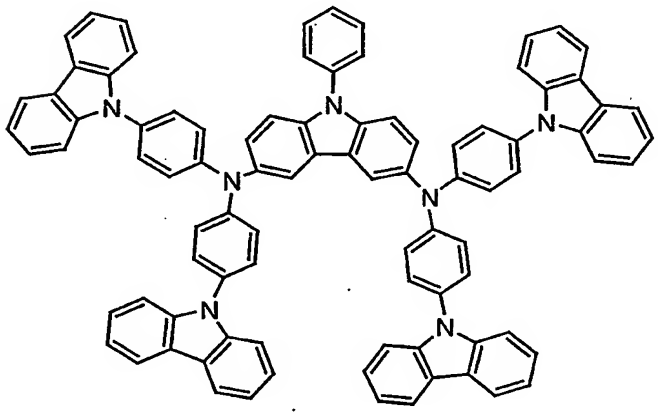

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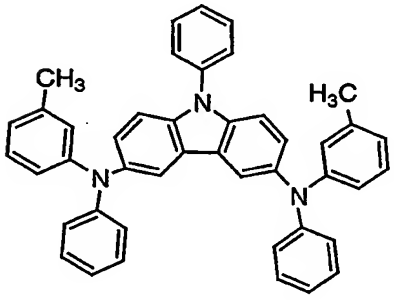
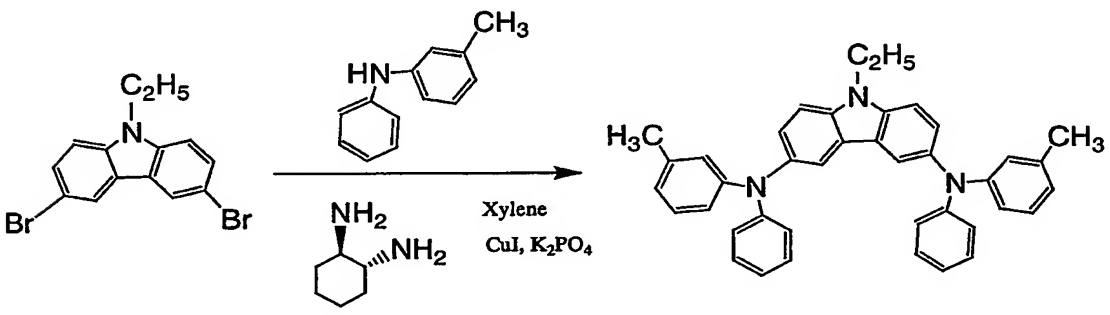


(75)

Various reactions can be applied to a synthesis method of the carbazole derivative according to the present invention. As a synthesis scheme of the carbazole derivative represented by the above-mentioned structure formula (56), there is the following method. However, the synthesis method of the carbazole derivative according to the present invention is not to be considered limited to this.



(76)

[Embodiment 2]

According to the present invention, a light-emitting element can be manufactured by using the carbazole derivative shown in Embodiment 1.

A light-emitting element according to the present invention has a structure that has an organic thin layer including a light-emitting layer (a layer including a luminescent material) between an anode and a cathode. The structure is not particularly limited, which can be selected appropriately for any purpose. For example, the structure may have protective films for suppressing quenching in a light-emitting region between the anode and the light-emitting layer or between the cathode and the light-emitting layer, that is, may have another layers such as a hole injecting layer, a hole transporting layer, an electron transporting layer, and a electron injecting layer. In this case, the carbazole derivative according to the present invention can be used as a

material for forming the light-emitting layer, the hole transporting layer, or the hole injecting layer.

In the light-emitting element according to the present invention, light generated by recombination of carriers in the layer including the luminescent material is emitted from one or both of the anode and the cathode to the outside. In other words, the anode is formed to include light-transmitting material in the case where light is emitted from the anode while the cathode is formed to include a light-transmitting material in the case where light is emitted from the cathode.

For the layer including the luminescent material, known materials can be used, and any of low molecular weight materials and high molecular weight materials can be used. The carbazole derivative according to the present invention is included in the layer including the luminescent material in the light-emitting element of the present embodiment. The materials for forming the layer including the luminescent material includes not only organic compounds but also an inorganic compound included in a portion of the layer including the luminescent material.

In the present invention, in the case of using the carbazole derivative for the light-emitting layer, the layer including the luminescent material can be formed by combining a light emitting layer including a luminescent material with a layer which do not include a luminescent material between electrodes. In other words, the layer including the luminescent material can have a laminated structure by combining layers such as the hole injecting layer, the hole transporting layer, the hole blocking layer, the electron transporting layer, and the electron injecting layer as appropriate in addition to the light-emitting layer.

Now, a light-emitting element that has a first electrode that functions as an anode; a hole injecting layer, a hole transporting layer, a light-emitting layer, an electron transporting layer, and a hole blocking layer, which are formed in this order on the first electrode; and a second electrode that functions as a cathode, which is formed further

thereon, and includes the carbazole derivative according to the present invention as a material for forming the hole transporting layer will be described.

It is preferable that the light-emitting element according to the present invention is supported by a substrate. The substrate is not particularly limited, and a substrate that is used for a conventional light-emitting element, for example, a substrate including a material such as glass, quartz, or transparent plastic can be used.

As an anode material for the light-emitting element according to the present invention, it is preferable to use a metal, an alloy, an electrically conductive compound, or a mixture thereof, which has a large work function (a work function of 4.0 eV or more). As a specific example of the anode material, a metal such as gold (Au), platinum (Pt), nickel (Ni), tungsten (W), chromium (Cr), molybdenum (Mo), iron (Fe), cobalt (Co), copper (Cu), or palladium (Pd), and a nitride of a metal material such as TiN can be used in addition to ITO (indium tin oxide) and IZO (indium zinc oxide) of indium oxide mixed with zinc oxide (ZnO) at 2 to 20 %.

On the other hand, as a cathode material, it is preferable to use a metal, an alloy, an electrically conductive compound, or a mixture of these, which has a small work function (a work function of 3.8 eV or less). As a specific example of the cathode material, an alkali metal (such as Li, Na, K, or Cs), an alkali earth metal (such as Mg or Ca), gold, silver, lead, aluminum, an alloy or mixed metal of aluminum and lithium, and an alloy or mixed metal of magnesium and silver can be used. Further, between the cathode including the metal mentioned above and an organic layer, one of a metal oxide and a metal halide may be used as an electron injecting layer. As specific examples of the electron injecting layer, metal oxides such as lithium oxide (Li_2O), magnesium oxide (MgO), and aluminum oxide (Al_2O_3) and metal halides such as lithium fluoride (LiF), magnesium fluoride (MgF_2), strontium fluoride (SrF_2) can be used.

A thin film including the anode material and a thin film including the cathode material are formed by a method such as evaporation or sputtering to form the anode

and the cathode respectively, which preferably have a film thickness of 10 to 500 nm.

As a hole injecting material for forming the hole injection layer, porphyrin-based compounds are efficient among organic compounds. For example, phthalocyanine (hereinafter, referred to as H_2-Pc) and copper phthalocyanine (hereinafter, referred to as Cu-Pc) can be used. In addition, a material of a chemically doped conductive polymer such as polyethylene dioxythiophene (hereinafter, referred to as PEDOT) doped with polystyrene sulfonate (hereinafter, referred to as PSS) can be used.

As a hole transporting material for forming the hole transporting layer, the carbazole derivative according to the present invention can be used. In this case, any of the carbazole derivatives represented by the structure formulas (2) to (75) may be used.

As a luminescent material to be used for the light-emitting layer, in addition to metal complexes such as Alq_3, $Almq_3$, $BeBq_2$, BAlq, $Zn(BOX)_2$, and $Zn(BTZ)_2$, various fluorescent pigments such as (hereinafter, referred to as TBiPy) are efficient. One of these materials may be used to form the light-emitting layer, or two or more of the materials may be combined to form the light-emitting layer.

As an electron transporting material for forming the electron transporting material, a metal complex that has a quinoline moiety or a benzoquinoline moiety such as tris (8 - quinolinolato) aluminum (hereinafter, referred to as Alq_3), tris (4 - methyl - 8 - quinolinolato) aluminum (hereinafter, referred to as $Almq_3$), or bis (10 - hydroxybenzo [h] - quinolinato) beryllium (herenafter, referred to as $BeBq_2$), and bis (2 - methyl - 8 - quinolinolato) - (4 - hydroxy - biphenylyl) - aluminum (hereinafter, referred to as BAlq) that is a mixed ligand complex are preferred. In addition, there is also a metal complex that has an oxazole-based, thiazole-based, or benzimidazole-based ligand such as bis [2 - (2 - hydroxyphenyl) - benzoxazolato] zinc (hereinafter, referred to as $Zn(BOX)_2$), bis [2 - (2 - hydroxyphenyl) - benzothiazolato] zinc (hereinafter, referred to

as $Zn(BTZ)_2$), or tris - (2 - (2' - hydroxyphenyl) - 1 - phenyl - 1H - benzimidazolate) aluminum (hereinafter, referred to as $Al(PBI)_3$). In addition to the metal complexe, oxadiazole derivatives such as 2 - (4 - biphenylyl) - 5 - (4 - tert - butylphenyl) - 1, 3, 4 - oxadiazole (hereinafter, referred to as PBD) and 1, 3 - bis [5 - (p - tert - butylphenyl) - 1, 3, 4 - oxadiazole - 2 - yl] benzene (hereinafter, referred to as OXD-7), triazole derivatives such as 3 - (4 - tert - butylphenyl) - 4 - phenyl - 5 - (4 - biphenylyl) - 1, 2, 4 - triazole (hereinafter, referred to as TAZ) and 3 - (4 - tert - butylphenyl) - 4 - (4 - ethylphenyl) - 5 - (4 - biphenylyl) - 1, 2, 4 - triazole (hereinafter, referred to as p-EtTAZ), phenanthroline derivatives such as bathophenanthroline (hereinafter, referred to as BPhen) and bathocuproin (hereinafter, referred to as BCP), and benzimidazole derivatives such as 2, 2', 2" - (1, 3, 5 - benzenetriyl) tris - [1 - phenyl - 1H - benzimidazole] (hereinafter, referred to as TPBI), 1, 3, 5 - tris [4 - (1 - phenyl - 1H - benzimidazole - 2 - yl) phenyl] benzene (hereinafter, referred to as TPBIBB), and 9 - phenyl - 2, 4, 5, 7 - tetrakis (1 - phenyl - 1H - benzimidazole - 2 - yl) - carbazole (hereinafter, referred to as PBIC) can be used.

As a hole blocking material for forming the hole blocking layer, materials such as the above-mentioned BAlq, OXD-7, TAZ, p-EtTAZ, BPhen, and BCP can be used.

As described above, the carbazole derivative according to the present invention can be used as a material for forming the hole transporting layer.

Next, a case of using the carbazole derivative according to the present invention as a material for forming the light-emitting layer will be described.

The carbazole derivative according to the present invention can also be used as a host material or guest material of the light-emitting layer.

In the case of using the carbazole derivative according to the present invention as a host material of the light-emitting layer, triplet luminescent materials (phosphorescent materials) such as tris (2 - phenylpyridine) iridium (hereinafter, referred to as $Ir(ppy)_3$) and 2, 3, 7, 8, 12, 13, 17, 18 - octaethyl - 21H, 23H - porphyrin -

platinum (hereinafter, referred to as PtOEP) can be used as a guest material in addition to quinacridone, diethyl quinacridone (DEQ), rubrene, perylene, DPT, Co-6, PMDFB, BTX, ABTX, DCM, and DCJT.

In the case of using the carbazole derivative according to the present invention as a guest material of the light-emitting layer, materials such as TPD, α-NPD, TCTA, PBD, OXD-7, BCP can be used as a host material.

As a hole transporting material for forming the hole transporting layer, an aromatic amine-based compound (that is, a compound that has a benzene ring-nitrogen bond) is preferred. As materials that are widely used, for example, in addition to N, N' - bis (3 - methylphenyl) - N, N' - diphenyl - [1, 1' - biphenyl] - 4, 4' - diamine (hereinafter, referred to as TPD), derivatives thereof such as 4, 4' - bis [N - (1 - naphthyl) - N - phenyl - amino] - biphenyl (hereinafter, referred to as α-NPD), starburst aromatic amine compounds such as 4, 4', 4" - tris (N-carbazolyl) - triphenylamine (hereinafter, referred to as TCTA), 4, 4', 4" - tris (N, N - diphenyl - amino) - triphenylamine (hereinafter, referred to as TDATA), and 4, 4', 4" - tris [N - (3 - methylphenyl) - N - phenyl - amino] - triphenylamine (hereinafter, referred to as MTDATA), and pyrene derivatives such as TBiPy are included.

As described above, a light-emitting element from which stable light emission can be obtained at a reduced driving voltage efficiently for a long stretch of time can be manufactured by using the carbazole derivative according to the present invention, which has material properties of having a great hole transporting property, permitting an uniform film to be formed, and being unlikely to undergo crystallization and morphologically stable.

[Embodiment 3]

In the present embodiment, a light-emitting element using the carbazole derivative according to the present invention is manufactured over a substrate including

a material such as glass, quartz, or transparent plastic. By manufacturing a plurality of light-emitting elements using the carbazole derivative according to the present invention over a substrate, a passive matrix light-emitting device can be manufactured. In addition, other than the substrate including the material such as glass, quartz, or transparent plastic, for example, as shown in Fig. 1, a light-emitting element in contact with a thin film transistor (TFT) array may be manufactured. In Fig. 1, reference numeral 10 denotes a substrate, portions 11 and 12 surrounded by dashed lines respectively denote TFTs, and reference numerals 14, 15, 16, and 17 respectively denote a first electrode, a layer including a luminescent material, a second electrode, and a wiring. In addition, a portion where the first electrode 14, the layer 15 including the luminescent material, and the second electrode 16 are laminated functions a light-emitting element 13. In this way, an active matrix light-emitting device where driving of a light-emitting element is controlled by a TFT can be manufactured. The structures of the TFTs are not particularly limited. For example, a staggered TFT and an inversely staggered TFT may be used. In addition, the degree of crystallinity a semiconductor layer forming the TFT is not particularly limited, either. A crystalline semiconductor layer or an amorphous semiconductor layer may be used to form the TFT.

[Example 1]

(Synthesis Example 1)

In Synthesis Example 1, an example of synthesizing the carbazole derivative represented by the above-mentioned structure formula (56) will be specifically exemplified.

In an atmosphere of argon, 16.59 g (30 mmol) of N - ethyl - 3, 6 - dibromocarbazole and 12.09 g (66 mmol) of 3 - methyldiphenylamine were dissolved in 100ml of dehydrated xylene. Then, 5.7 g (30 mmol) of copper iodide and 22.8 g (200

mmol) of trans-cyclohexanediamine were added to the xylene solution, and stirring was performed at 160 °C for 30 minutes. After the stirring, 27.6 g (130 mmol) of potassium phosphate was added, and stirring was performed further for 9 days. After the stirring and cooling to room temperature, 300 ml of toluene was added, and a precipitated object was filtered. The obtained filtrate was condensed, diethyl ether was added to the filtrate, and a precipitated object was filtered. Then, when methanol was added to the obtained filtrate, a tarry object was precipitated on a surface of the beaker. The filtrate with the tarry object was left at rest overnight, and the liquid phase was removed by decantation to obtain the tarry object. This obtained tarry object was purified by silica gel column chromatography using hexane:chloroform (1:2) to obtain 3, 6 - bis [N - (3 - methylphenyl) - N - phenylamino] - 9 - ethylcarbazole (the above structure formula (56); hereinafter, referred to as EtCzmP2) that is light green powder. The obtained EtCzmP2 was purified by sublimation while setting at a higher temperature of 300 °C and a lower temperature of 200 °C. The yield after the purification by sublimation was approximately 10 %.

It was determined according to a thermogravimetry-differential thermal analysis (TG-DTA) measurement that the thermal decomposition temperature of the obtained EtCzmP2 was 310 °C. When vacuum deposition was used to deposit the carbazole derivative, it was possible to form an uniform film.

When fluorescence spectrums of a thin film and solution (solvent: methanol) of the obtained EtCzmP2 were measured, the obtained fluorescence spectrums respectively had a maximum peak at 435 nm with respect to an excitation wavelength (312 nm) in the case of the thin film and a maximum peak at 400 nm with respect to an excitation wavelength (290 nm) in the case of the solution (Fig. 2). In addition, when UV-Vis absorption spectrums of the thin film and solution (solvent: methanol) of the obtained EtCzmP2 were measured, a maximum absorption wavelength of 312 nm was obtained in the case of the thin film and a maximum absorption wavelength of 303 nm was

obtained in the case of the solution (Fig. 3).

Further, the value of a HOMO level that was measured by using Electron Spectrometer for Chemical Analysis AC-2 (from Riken Keiki Co., Ltd.) is -5.18 eV. In addition, the value of a LUMO level that was estimated by adding the value of an absorption edge of the absorption spectrum (Fig. 3), as an energy gap, to the value of the HOMO level is -1.71 eV.

(Synthesis Example 2)

In Synthesis Example 2, an example of synthesizing the carbazole derivative represented by the above-mentioned structure formula (75) will be specifically exemplified.

In Synthesis Example 2, a raw material, 12.03 g (30 mmol) of N - phenyl - 3, 6 - dibromocarbazole was used to obtain 3, 6 - bis [N - (3 - methylphenyl) - N - phenylamino] - 9 - phenylcarbazole (the above structure formula (75); hereinafter, referred to as PhCzmP2) that is a carbazole derivative according to the present invention in the same way as in Synthesis Example 1 described above. The obtained PhCzmP2 was purified by sublimation while setting at a higher temperature of 290 °C and a lower temperature of 90 °C. The yield after the purification by sublimation was approximately 10 %. Fig. 4 shows a ^{1}H NMR spectrum of the obtained PhCzmP2, and Fig. 5 shows an enlarged view of a region (A) surrounded by a dashed line in Fig. 4. Here is data of the ^{1}H NMR of the obtained PhCzmP2.

^{1}H NMR (300 MHz, DMSO-d); δ = 3.31 (s, 6H), 6.74 (s, 2H), 6.75 (d, j = 6.0, 4H), 6.85-6.91 (m, 6H), 7.08 (t, j = 7.8, 2H), 7.15-7.20 (m, 6H), 7.33 (d, j = 8.7, 2H), 7.51 (t, j = 7.2, 1H), 7.58-7.68 (m, 4H), 7.91 (s, 2H)

It was determined according to a TG-DTA measurement that the thermal decomposition temperature of the obtained PhCzmP2 was 230 °C. When vacuum deposition was used to deposit the carbazole derivative, it was possible to form an

uniform film.

When fluorescence spectrums of a thin film and solution (solvent: methanol) of the obtained PhCzmP2 were measured, the obtained fluorescence spectrums respectively had a maximum peak at 430 nm with respect to an excitation wavelength (308 nm) in the case of the thin film and a maximum peak at 439 nm with respect to an excitation wavelength (320 nm) in the case of the solution (Fig. 6). In addition, when an UV-Vis absorption spectrum of the thin film of the obtained PhCzmP2 was measured, a maximum absorption wavelength of 308 nm was obtained (Fig. 7).

Further, the values of a HOMO level and a LUMO level that were measured in the same way as in Synthesis Example 1 described above are -5.40 eV and -2.39 eV, respectively.

(Synthesis Example 3)

In Synthesis Example 3, an example of synthesizing the carbazole derivative represented by the above-mentioned structure formula (76) will be specifically exemplified.

In Synthesis Example 3, raw materials, 12.03 g (30 mmol) of N - phenyl - 3, 6 - dibromocarbazole and diphenylamine (12.18 g, 72 mmol) were used to obtain N - phenyl - 3, 6 - di - (diphenylamino) carbazole (3, 6 - di - (diphenylamino) N - phenylcarbazole) (the above structure formula (76); hereinafter, referred to as PhCzP2) that is a carbazole derivative according to the present invention in the same way as in Synthesis Example 1 described above. The obtained PhCzP2 was purified by sublimation while setting at a higher temperature of 270 °C and a lower temperature of 175 °C. The yield after the purification by sublimation was approximately 50 %. Fig. 8 shows a ^{1}H NMR spectrum of the obtained PhCzP2, Fig. 9 shows a ^{13}C NMR spectrum of the obtained PhCzP2, and Fig. 10 shows an enlarged view of a region (A) surrounded by a dashed line in Fig. 9. Here is data of ^{1}H NMR and ^{13}C NMR of the

obtained PhCzP2.

^{1}H NMR (300 MHz, DMSO-d); δ = 6.88-6.95 (m, 12H), 7.17-7.23 (m, 10H), 7.35 (d, j = 4.5, 6H), 7.56-7.69 (m, 5H), 7.97 (s, 2H)

^{13}C NMR (300 MHz, DMSO-d); δ = 111.1, 119.4, 121.6, 122.0, 123.6, 126.4, 126.8, 127.8, 129.3, 130.2, 136.7, 138.2, 139.9, 148.0

It was determined according to a TG-DTA measurement that the thermal decomposition temperature of the obtained PhCzP2 was 365 °C. When vacuum deposition was used to deposit the carbazole derivative, it was possible to form an uniform film.

When fluorescence spectrums of a thin film and solution (solvent: dichloromethane) of the obtained PhCzP2 were measured, the obtained fluorescence spectrums respectively had a maximum peak at 429 nm with respect to an excitation wavelength (313 nm) in the case of the thin film and a maximum peak at 435 nm with respect to an excitation wavelength (315 nm) in the case of the solution (Fig. 11). In addition, when UV-Vis absorption spectrums of the thin film and dichloromethane solution of the obtained PhCzP2 were measured, a maximum absorption wavelength of 313 nm was obtained in the case of the thin film and a maximum absorption wavelength of 305 nm was obtained in the case of the solution (Fig. 12).

Further, the values of a HOMO level and a LUMO level that were measured in the same way as in Synthesis Example 1 described above are -5.31 eV and -2.57 eV, respectively.

[Example 2]

In the present example, a case of using a carbazole derivative according to the present invention for a portion of a layer including a luminescent material to manufacture a light-emitting element, specifically, a structure in the case of a carbazole derivative according to the present invention as a hole transporting material will be

described with reference to Fig. 13.

First, a first electrode 101 for a light-emitting element is formed over a substrate 100. In the present example, the first electrode 101 functions as an anode. ITO that is a transparent conductive film is used as a material to form the first electrode 101 with a film thickness of 110 nm by sputtering.

Next, a layer 102 including a luminescent material is formed on the first electrode (anode) 101. The layer 102 including the luminescent material in the present example has a laminated structure including a hole injecting layer 111, a hole transporting layer 112, a light-emitting layer 113, an electron transporting layer 114, and an electron injecting layer 115.

The substrate over which the first electrode 101 is formed fixed in a substrate holder of a commercially produced vacuum deposition device with the surface at which the first electrode 101 is formed down, copper phthalocyanine (hereinafter, referred to as Cu-Pc) is put in an evaporation source provided in the vacuum deposition device, and then, the hole injecting layer 111 is formed by evaporation using resistance heating to have a film thickness of 20 nm. As a material for forming the hole injecting layer 111, a known hole injecting material can be used.

Then, the hole transporting layer 112 is formed. In the present example, as a material for forming the hole transporting layer 112, EtCzmP2 that is a carbazole derivative according to the present invention is used to form the hole transporting layer 112 with a film thickness of 40 nm by evaporation.

Then, the light-emitting layer 113 is formed. In the light-emitting layer 113, a hole and an electron are recombined to generate luminescence (to emit light). As a material for forming the light-emitting layer 113, known luminescent materials can be used. In the present example, TBiPy is used to form the light-emitting layer 113 with a film thickness of 30 nm by evaporation.

Then, the electron transporting layer 114 is formed. As a material for forming

the electron transporting layer 114, a known electron transporting material can be used. In the present example, Alq_3 is used to form the electron transporting layer 114 with a film thickness of 20 nm by evaporation.

Then, the electron injecting layer 115 is formed. As a material for forming the electron injecting layer 115, a known electron injecting material can be used. In the present example, calcium fluoride (hereinafter, referred to as CaF_2) is used to form the electron transporting layer 115 with a film thickness of 2 nm by evaporation.

After forming the layer 102 including the luminescent material, which is formed by laminating the hole injecting layer 111, the hole transporting layer 112, the light-emitting layer 113, the electron transporting layer 114, and the electron injecting layer 115 in this way, a second electrode 103 that functions as a cathode is formed by sputtering or evaporation. In the present example, aluminum (150 nm in film thickness) is formed by evaporation on the layer 102 including the luminescent material to obtain the second electrode 103.

In this way, the light-emitting element using the carbazole derivative according to the present invention is formed.

When a voltage is applied to the manufactured light-emitting element, blue luminescence was observed at a voltage of 5 V or more, and at an applied voltage of 10 V, blue luminescence with a luminance of 949 cd/m^2 (CIE chromatic coordinated of the EL (electroluminescence) : x = 0.170, y = 0.184) was observed. The luminous efficiency at the voltage of 10 V was 1.49 cd/A.

[Example 3]

In the present example, a case of using a carbazole derivative according to the present invention as a hole transporting material will be described with reference to Fig. 14. In the present example, structures of a substrate 200, a first electrode 201, a second electrode 203, a hole injecting layer 211, a light-emitting layer 213, an electron

transporting layer 214, and an electron injecting layer 216 are the same as those of Example 2. Accordingly, descriptions thereof are omitted.

Of a layer 202 including a luminescent material to be formed on a first electrode 201, a hole transporting layer 212 to be formed to come in contact with the hole injecting layer 211 as shown in Fig. 14 is formed by evaporation using PhCzmP2 that is a carbazole derivative according to the present invention to have a film thickness of 40 nm.

Then, a light-emitting element using the carbazole derivative according to the present invention can be manufactured by forming the second electrode 203 on the layer 202 including the luminescent material, which is formed by laminating the hole injecting layer 211, the hole transporting layer 212, the light-emitting layer 213, the electron transporting layer 214, and the electron injecting layer 215 in this way.

In this way, the light-emitting element using the carbazole derivative according to the present invention is manufactured.

When a voltage is applied to the manufactured light-emitting element, blue luminescence was observed at a voltage of 4.6 V or more, and at an applied voltage of 10 V, blue luminescence with a luminance of 2105 cd/m^2 (CIE chromatic coordinated of the EL (electroluminescence) : x = 0.197, y = 0.239) was observed. The luminous efficiency at the voltage of 10 V was 1.87 cd/A.

The carbazole derivative according to the present invention has a hole transporting property. Therefore, the carbazole derivative can be used as a hole transporting layer of a layer including a luminescent material, as shown in Examples 2 and 3 described above. Further, the carbazole derivative according to the present invention permits an uniform film to be formed, and is unlikely to undergo crystallization and morphologically stable, which makes it possible to expand the life of a light-emitting element.

[Example 4]

In the present example, a light-emitting device that has a light-emitting element according to the present invention in a pixel portion will be described with reference to Figs. 15A and 15B. Fig. 15A is a top view showing the light-emitting device and Fig. 15B is a cross-sectional view taken along line A-A' in Fig. 15A. Reference numeral 601 indicated by a dotted line denotes a driver circuit portion (a source side driver circuit), reference numeral 602 denotes a pixel portion, and reference numeral 603 denotes a driver circuit portion (a gate side driver circuit). In addition, reference numerals 604 and 605 denote a sealing substrate and a sealing material, respectively. The inside surrounded by the sealing material 605 is a space 607.

Reference numeral 608 denotes a leading wiring for transmitting signals to be input to the source side driver circuit 601 and the gate side driver circuit 603, and receives signals such as a video signal, a clock signal, a start signal, and a reset signal from FPC (Flexible Printed Circuit) 609 that serves as an external input terminal. A printed wiring board (PWB) may be attached to this FPC. The light-emitting device in the specification includes not only a light-emitting device body but also a state where an FPC or a PWB is attached thereto.

Next, the sectional structure will be explained with reference to Fig. 15B. The driver circuits and the pixel portion are formed over a substrate 610. Here, the source side driver circuit 601 as the driver circuit portion and the pixel portion 602 are shown.

In the source side driver circuit 601, a CMOS circuit is formed of a combination of an n-channel TFT 623 and a p-channel TFT 624. The TFTs forming the driver circuit may be formed of a known CMOS circuit, PMOS circuit, or NMOS circuit. Although the present example shows a driver integrated type in which a driver circuit is formed over a substrate, which is not always necessary, the driver circuit can be formed not over the substrate but outside the substrate.

The pixel portion 602 has a plurality of pixels, each including a switching TFT

611, a current controlling TFT 612, and a first electrode 613 electrically connected to a drain of the controlling TFT 612. In addition, an insulator 614 is formed to cover an edge of the first electrode 613. Here, a positive photosensitive acrylic resin film is used to form the insulator 614.

Besides, in order to obtain a favorable coverage, the insulator 614 is made to have a top portion or bottom potion formed with a curved surface with a curvature. For example, in the case of using positive photosensitive acrylic as a material for the insulator 614, it is preferable that only a top portion of the insulator 614 has a curved surface with a curvature radius (0.2 μm to 3 μm). In addition, any of a negative photosensitive material that becomes insoluble in an etchant by light and a positive photosensitive material that becomes soluble in an etchant by light can be used as the insulator 614.

On the first electrode 613, an electroluminescent layer which is a layer including a luminescent material 616 and a second electrode 617 are formed. Here, as a material to be used for the first electrode 613 that functions as an anode, it is preferable to use a material that has a large work function. For example, in addition to single layers such as an ITO (indium tin oxide) film, an indium zinc oxide (IZO) film, a titanium nitride film, a chromium film, a tungsten film, a Zn film, and a Pt film, laminated structures such as a laminate of a titanium nitride film and a film including aluminum as its main component and a three-layer structure of a titanium nitride film, a film including aluminum as its main component, and a titanium nitride film can be used. When a laminated structure is employed, the wiring has a lower resistance, favorable ohmic contact can be taken, and it is possible to function as an anode.

The electroluminescent layer 616 is formed by evaporation that uses an evaporation mask or by inkjet. The electroluminescent layer 616 includes a carbazole derivative according to the present invention. As a material to be used in combination with the carbazole derivative, low molecular weight materials, middle molecular weight

materials (including an oligomer and a dendrimer) or high molecular weight materials may be used. In addition, as a material to be used for the layer including the luminescent material, it is often the case that an organic material is used for a single layer or laminate. However, the present invention includes a structure in which an inorganic compound is used for a part of a film including an organic compound.

In addition, as a material to be used for the second electrode (cathode) 617 formed on the electroluminescent layer 616, a material that has a small work function (Al, Ag, Li, or Ca; an alloy thereof such as MgAg, MgIn, AlLi, or CaF_2; or CaN) may be used. In the case of transmitting light generated in the electroluminescent layer 616 through the second electrode 617, it is preferable to use a laminate of a metal thin film that has a thinned film thickness and a transparent conductive film (such as ITO (an alloy of indium oxide and tin oxide), an ally of indium oxide and zinc oxide (In_2O_3-ZnO), or zinc oxide (ZnO)) as the second electrode (cathode) 617.

Further, the sealing substrate 604 and the substrate 610 are bonded with the sealing material 605 to have a structure where a light-emitting element 618 is provided in the space 607 surrounded by the substrate 610, the sealing substrate 604, and the sealing material 605. The space 607 also includes a structure of filling with the sealing material 605 in addition to a case of filling with an inert gas (such as nitrogen or argon).

It is preferable to use an epoxy resin for the sealing material 605. In addition, it is desirable to use a material that hardly allows permeation of moisture or oxygen. Further, as a material to be used for the sealing substrate 604, a plastic substrate including FRP (Fiberglass-Reinforced Plastics), PVF (polyvinylfluoride), Mylar, polyester, or acrylic can be used besides a glass substrate and a quarts substrate.

In this way, the light-emitting device that has the light-emitting element according to the present invention can be obtained.

The light-emitting device shown in the present example can be implemented freely in combination with any of the structures of the light-emitting elements shown in

Examples 1 to 3.

[Example 5]

In the present example, various electronic devices completed by using a light-emitting device that has a light-emitting element according to the present invention will be described.

As examples of electronic devices equipped with a light-emitting device formed according to the present invention, a video camera, a digital camera, a goggle-type display (head mount display), a navigation system, a sound reproduction device (such as an in-car audio system or an audio set), a laptop personal computer, a game machine, a personal digital assistance (such as a mobile computer, a cellular phone, a portable game machine, or an electronic book), and an image reproduction device equipped with a recording medium (specifically, a device equipped with a display device, which can reproduce a recording medium such as a digital versatile disc (DVD) and display the image) can be given. Figs. 16A and 16B show some specific examples of these electronic devices, which will be described.

Figs. 16A and 16B are a display device, which includes a frame body 2001, a support 2002, a display portion 2003, a speaker portion 2004, and a video input terminal 2005. A light-emitting device formed according to the present invention is used for the display portion 2003 to manufacture the display device. The display device includes all devices for displaying information such as for a personal computer, for receiving TV broad casting, and for displaying an advertisement.

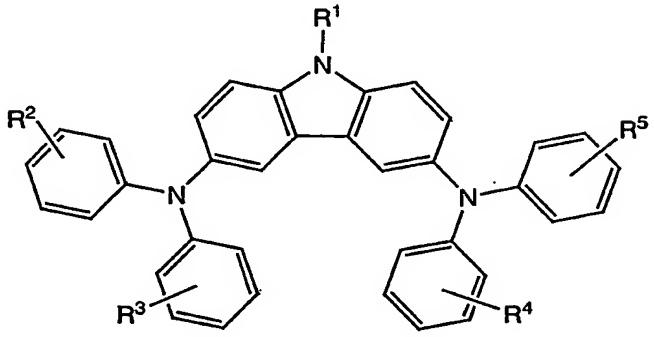
Fig. 16C is a cellular phone, which includes a main body 2701, a frame body 2702, a display portion 2703, a voice input portion 2704, a voice output portion 2705, an operation key 2706, an external connection port 2707, and an antenna 2708. A light-emitting device that has a light-emitting element according to the present invention is used for the display portion 2703 to manufacture the cellular phone.

As described above, a light-emitting device that has a light-emitting element according to the present invention is quite widely applied. In addition, since a carbazole derivative according to the present invention is used to form the light-emitting element that is used for the light-emitting device, the light-emitting element has features of a low driving voltage and a long lifetime. Therefore, it is possible to reduce power consumption and extend a lifetime (that is, favorable display images can be obtained for a long time) by applying this light-emitting device to electronic devices in all fields.

Although the present invention has been fully described by way of example with reference to the accompanying drawings, it is to be understood that various changes and modifications will be apparent to those skilled in the art. Therefore, unless such changes and modifications depart from the scope of the present invention hereinafter defined, they should be construed as being included therein.

CLAIMS

1. A carbazole derivative represented by the following general formula (1),



(1)

wherein R_1 is one of hydrogen, halogen, a cyano group, an alkyl group having 1 to 20 carbon atoms, a haloalkyl group having 1 to 20 carbon atoms, an alkoxyl group having 1 to 20 carbon atoms, a substituted or unsubstituted aryl group, and a substituted or unsubstituted heterocyclic group, and R_2 to R_5 are identical or different, and are individually one of hydrogen, halogen, a cyano group, an alkyl group having 1 to 20 carbon atoms, an alkoxyl group having 1 to 20 carbon atoms, an acyl group having 1 to 20 carbon atoms, a haloalkyl group having 1 to 20 carbon atoms, a dialkylamino group having 1 to 20 carbon atoms, a diarylamino group having 1 to 20 carbon atoms, a substituted or unsubstituted heterocyclic group, and a carbazolyl group.

2. A light-emitting element comprising the carbazole derivative according to claim 1.

3. A light-emitting element comprising the carbazole derivative according to claim 1 as a hole transporting material.

4. A light-emitting element comprising the carbazole derivative according to claim 1 as a luminescent material.

5. A light-emitting element comprising the carbazole derivative according to claim 1 and a guest material.

6. A light-emitting element comprising the carbazole derivative according to claim 1 and a host material.

7. A light-emitting device comprising the carbazole derivative according to claim 1.

8. An electronic device comprising the carbazole derivative according to claim 1, wherein the electronic device is selected from the group consisting of a video camera, a digital camera, a goggle-type display, a navigation system, a sound reproduction device, a laptop personal computer, a game machine, a personal digital assistance and an image reproduction device equipped with a recording medium.

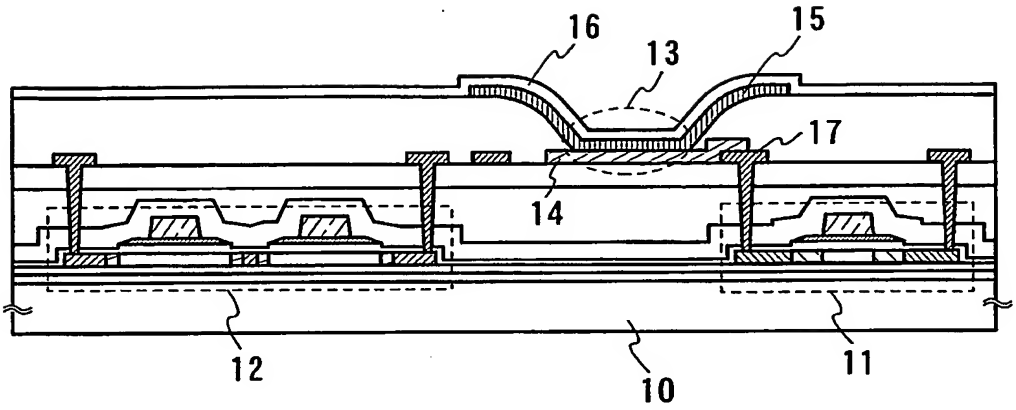


Fig. 1

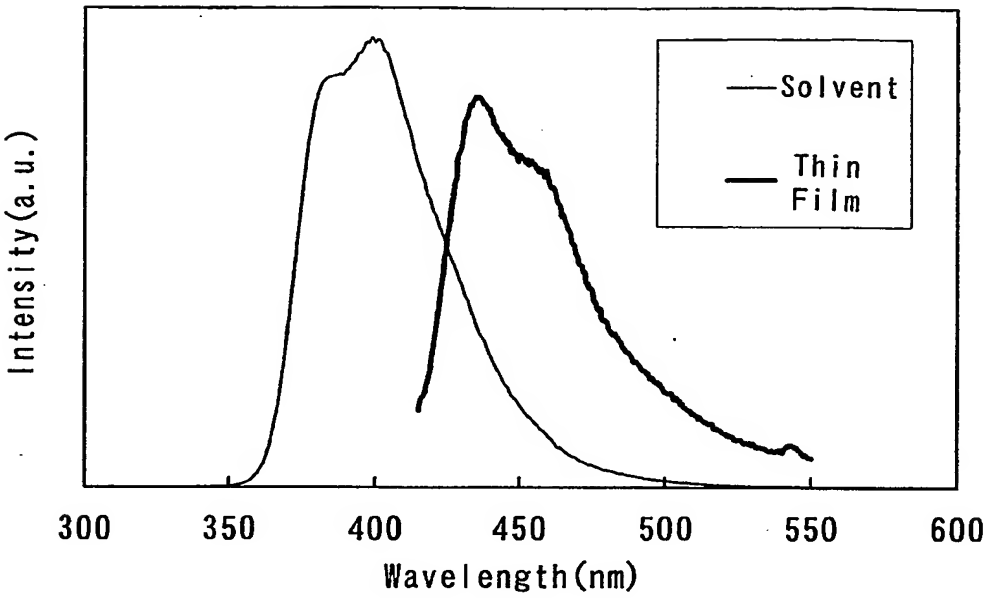

Solvent
Thin Film
Intensity(a.u.)
300
350
400
450
500
550
600
Wavelength(nm)

Fig. 2

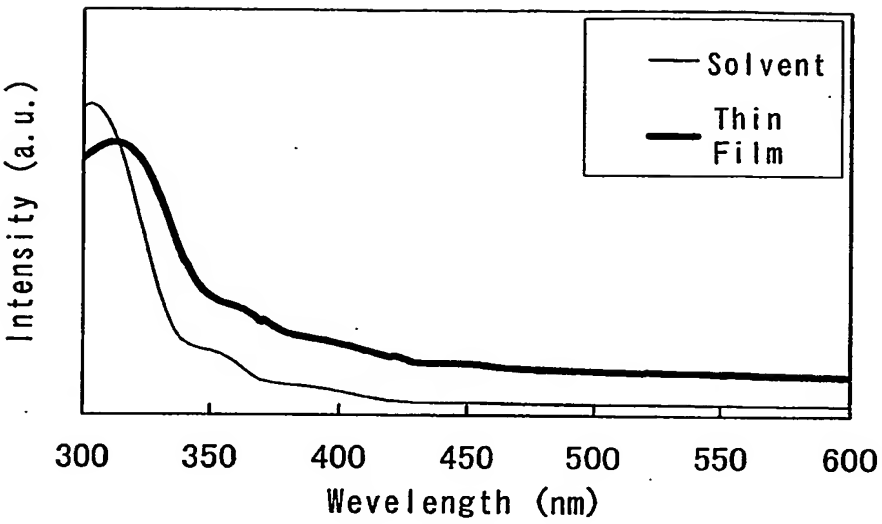

Solvent
Thin Film
Intensity (a.u.)
300
350
400
450
500
550
600
Wevelength (nm)

Fig. 3

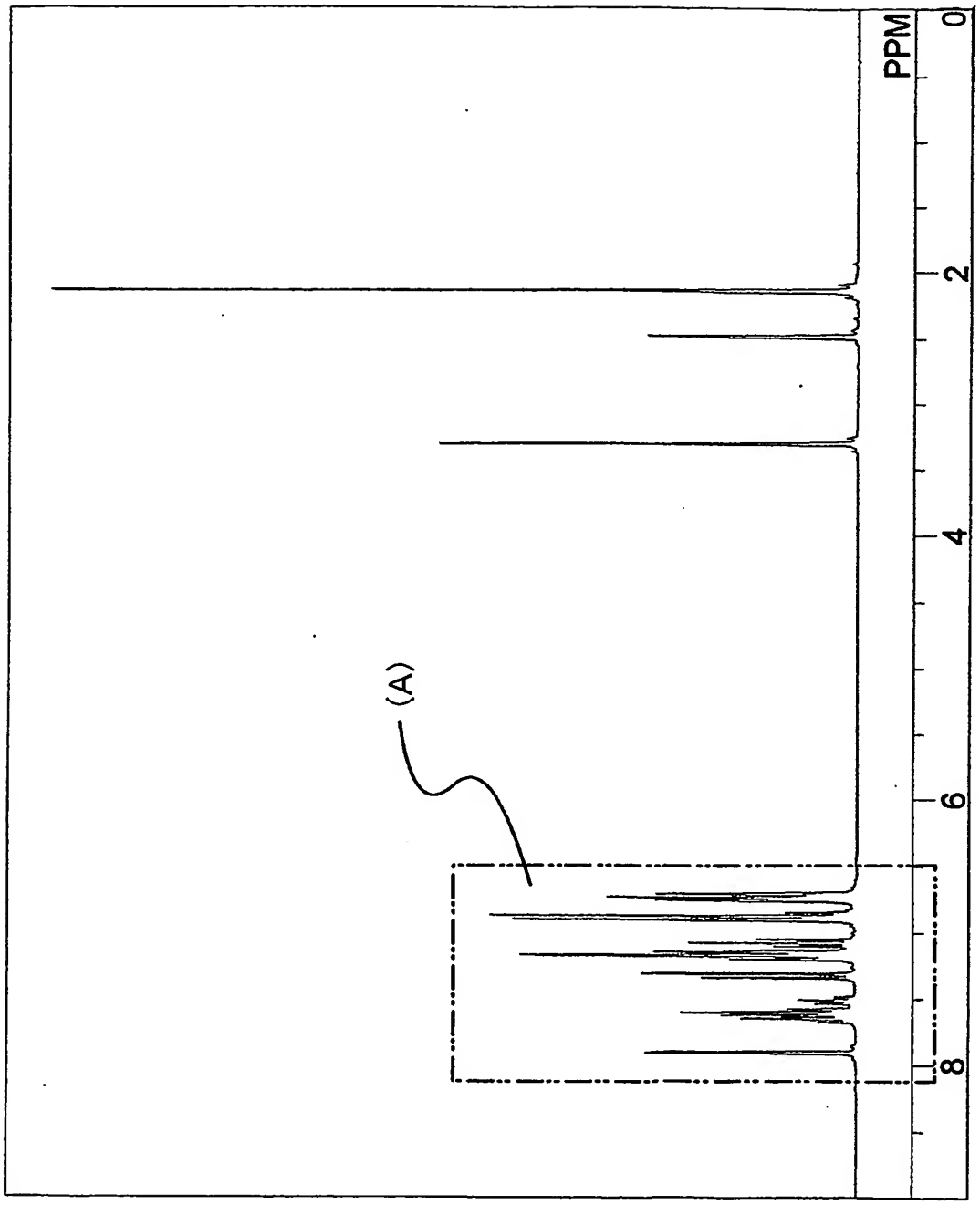


Fig. 4

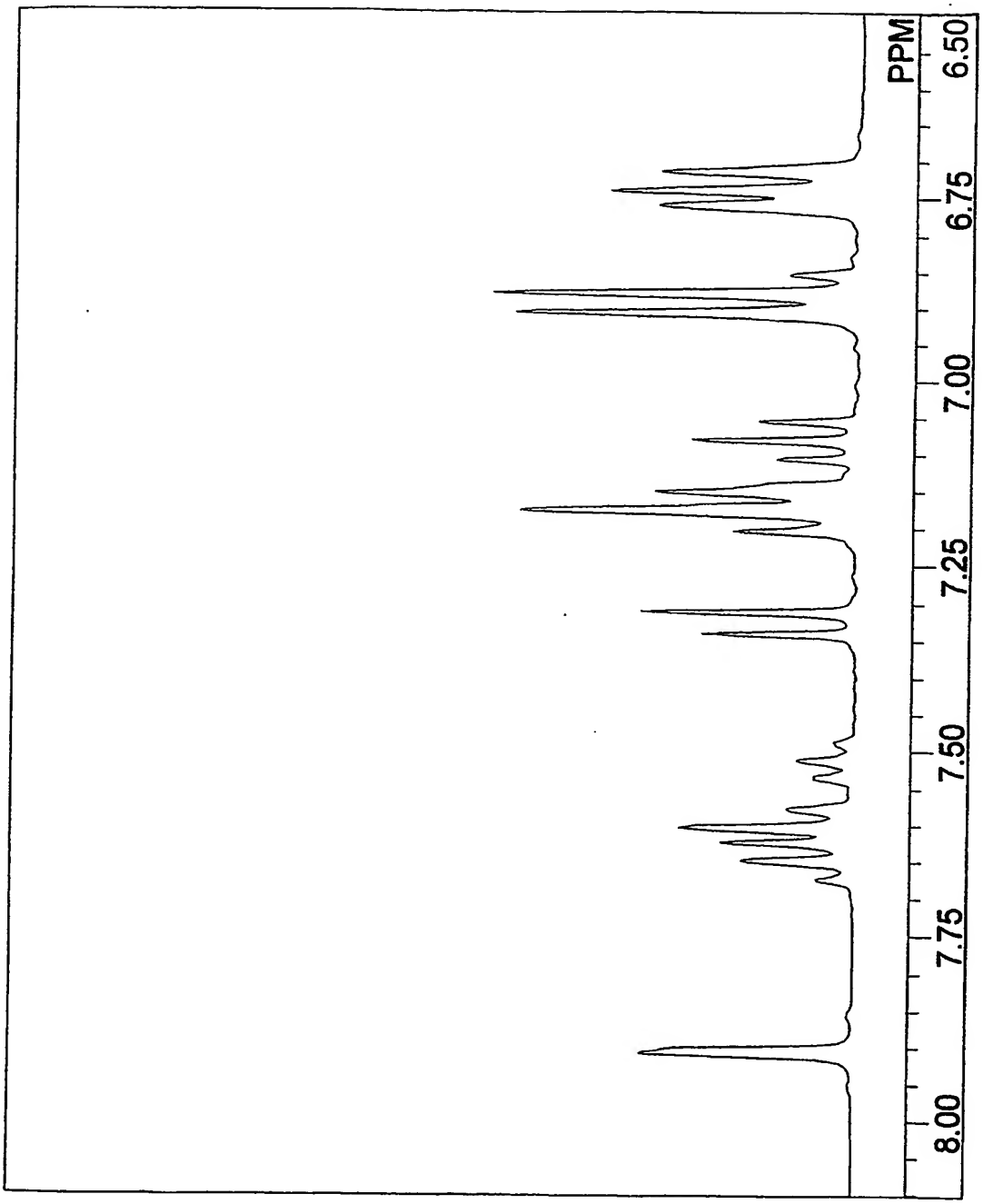


Fig. 5

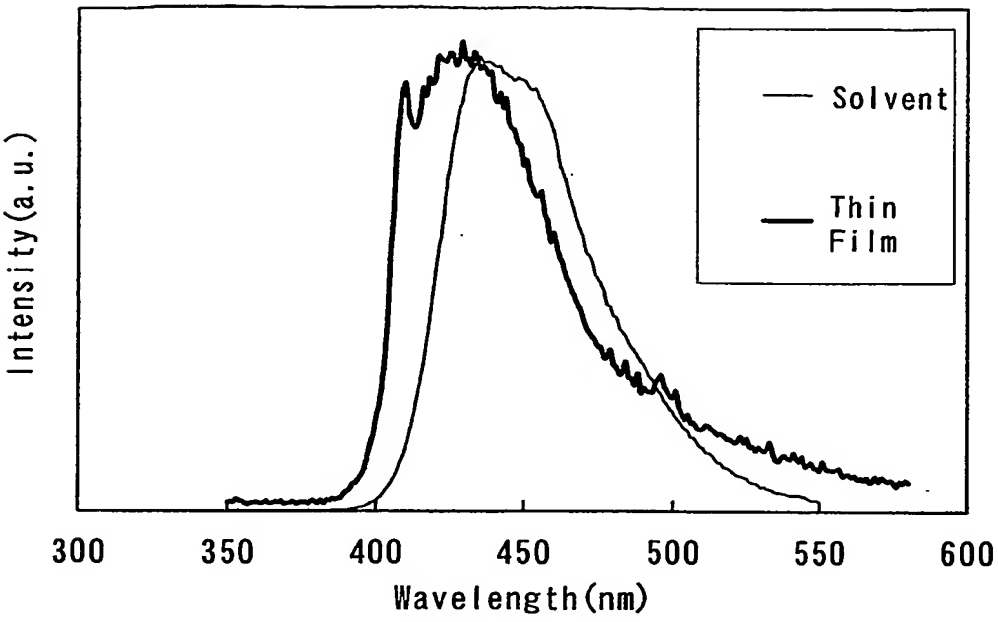

Solvent
Thin Film
Intensity(a.u.)
300
350
400
450
500
550
600
Wavelength(nm)

Fig. 6

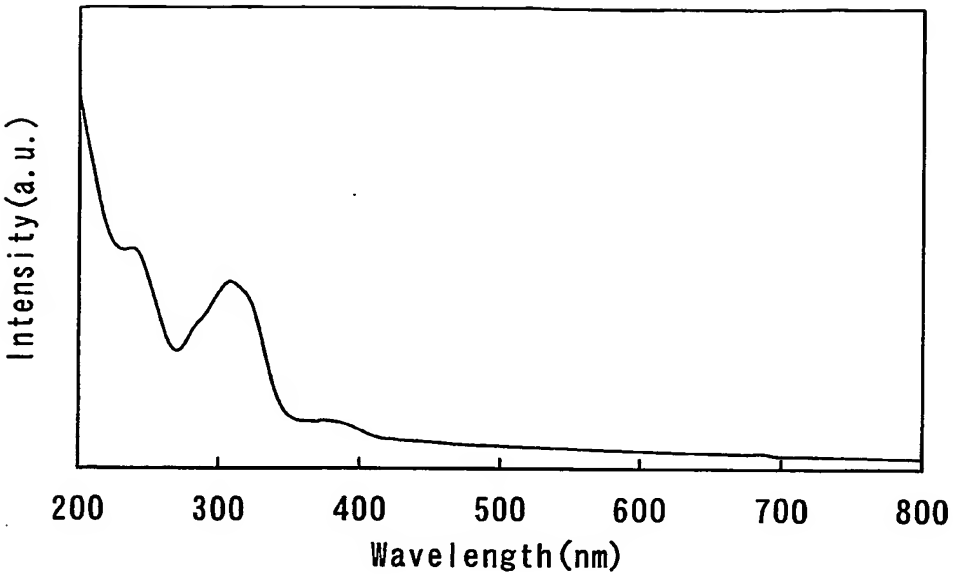


Fig. 7

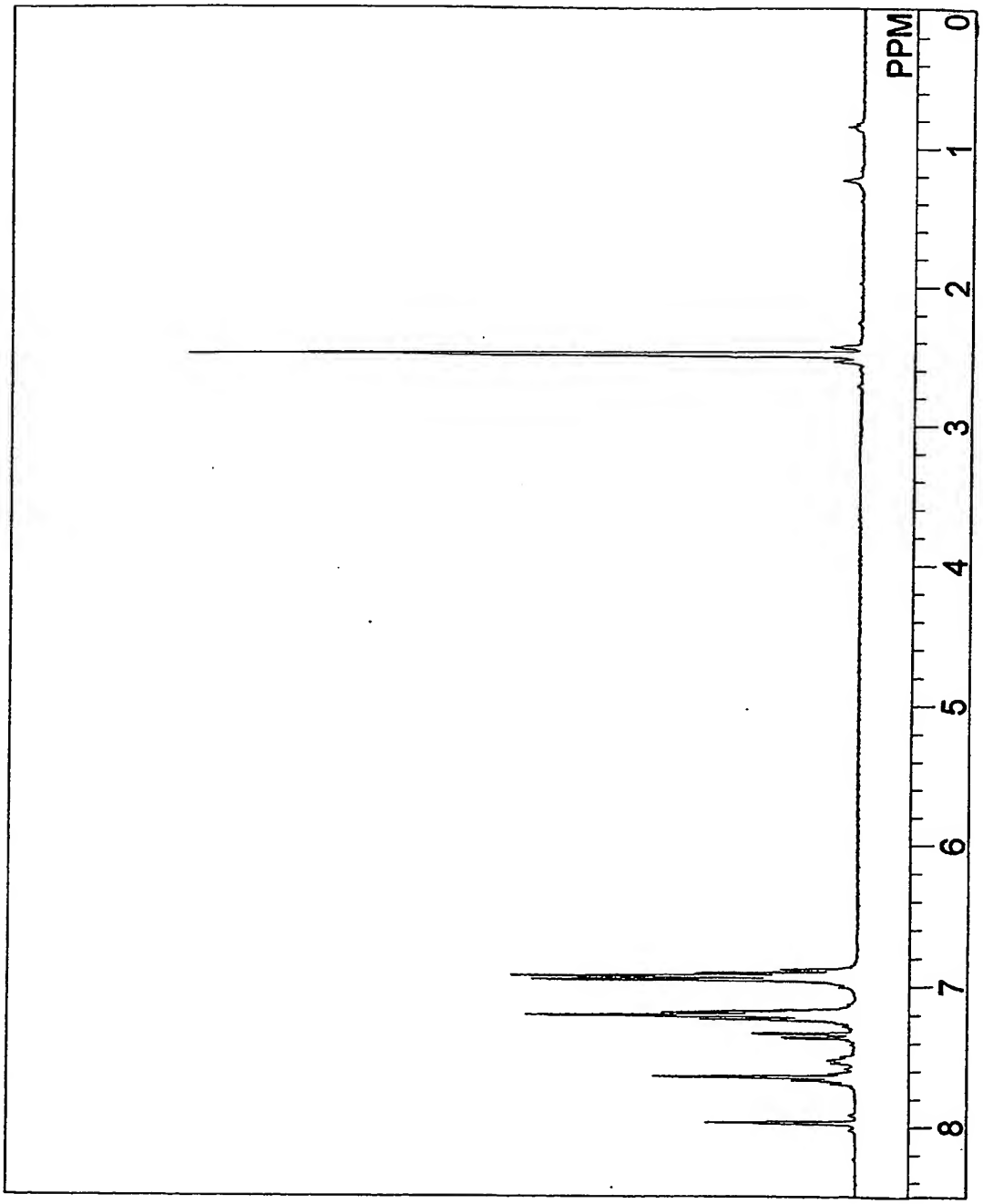


Fig. 8

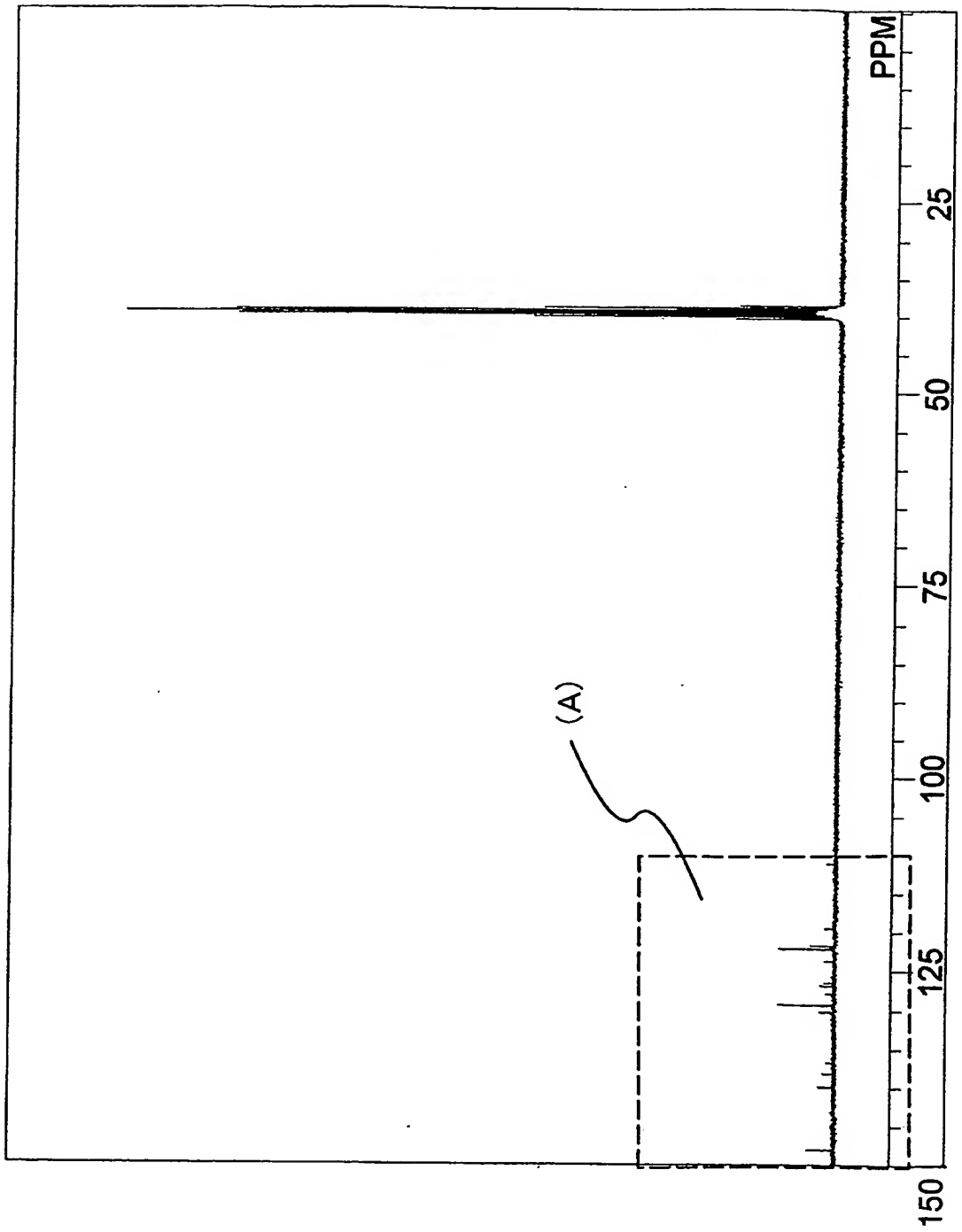


Fig. 9

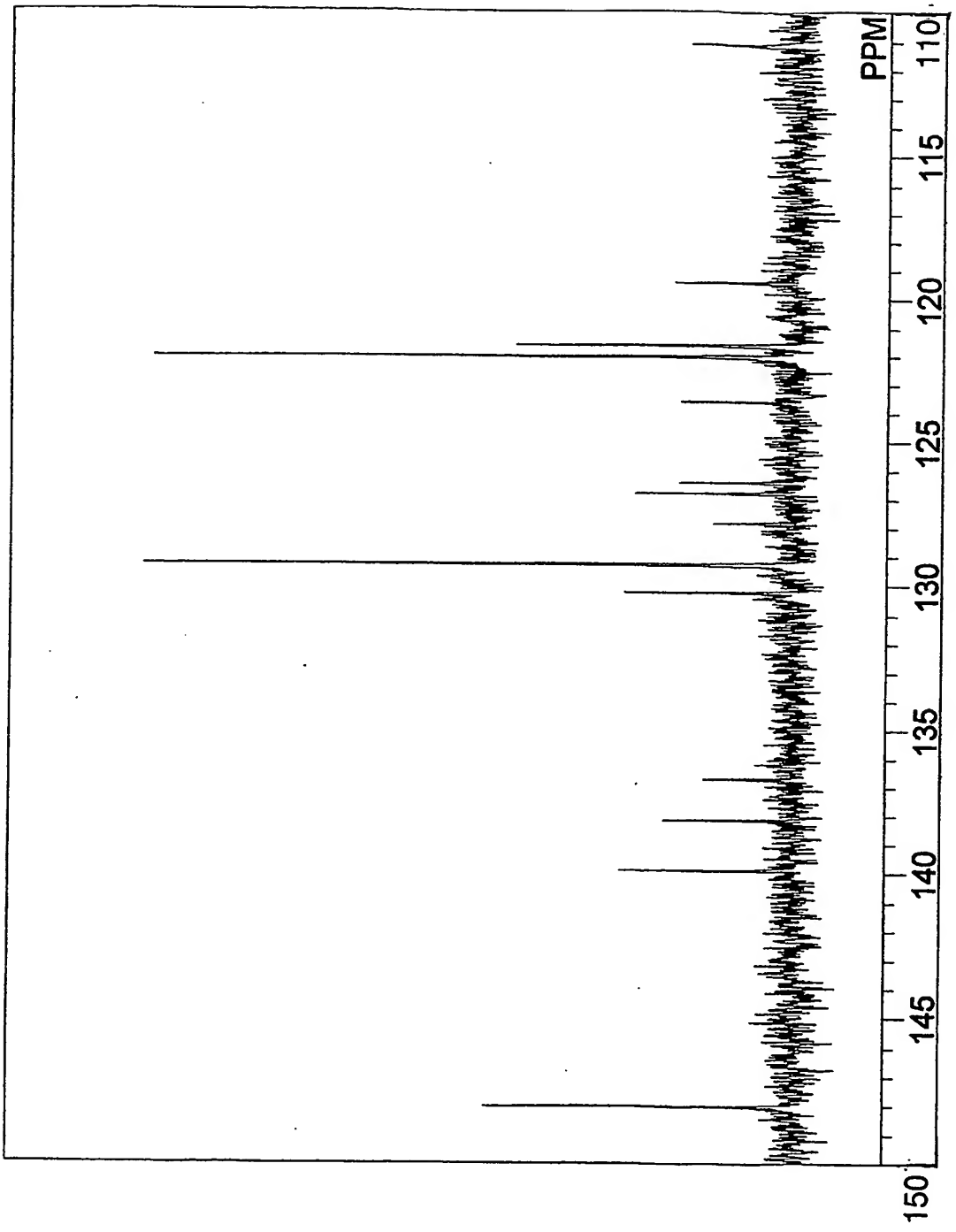


Fig. 10

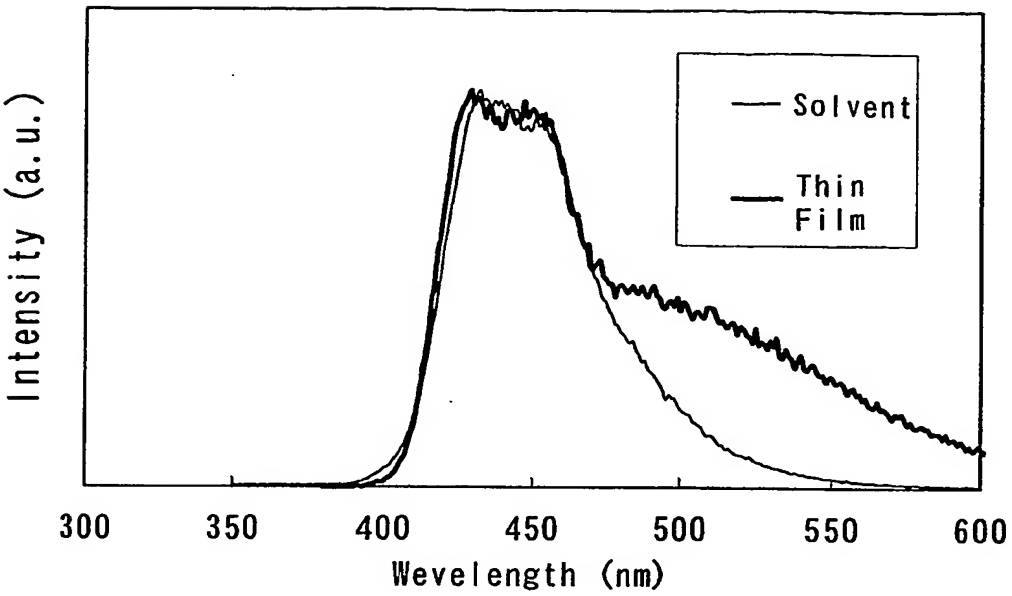

Intensity (a.u.)
Solvent
Thin Film
300
350
400
450
500
550
600
Wevelength (nm)

Fig. 11

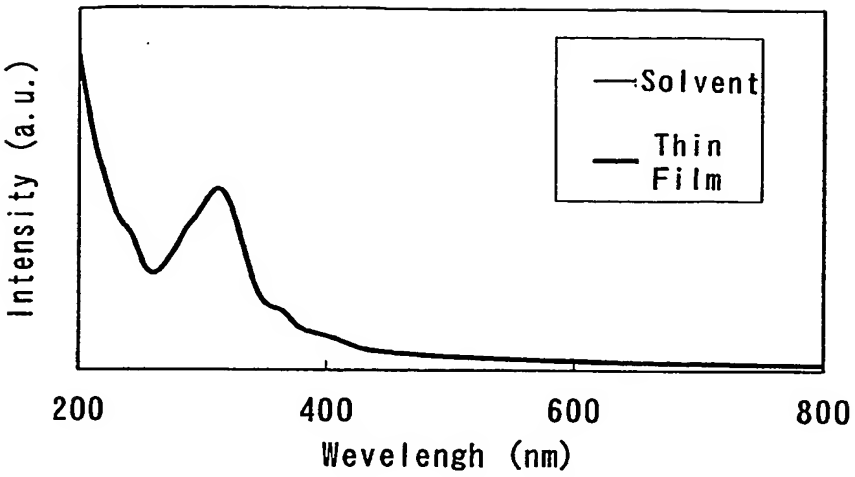


Fig. 12

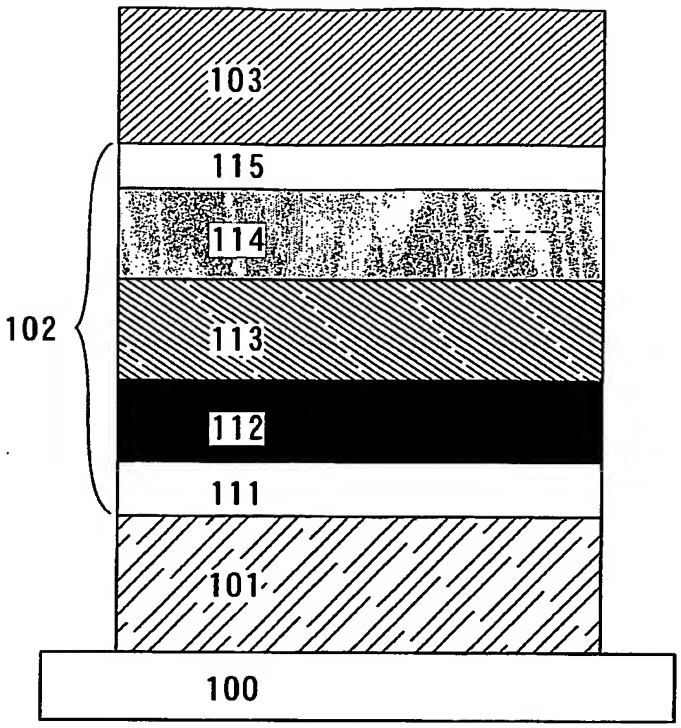


Fig. 13

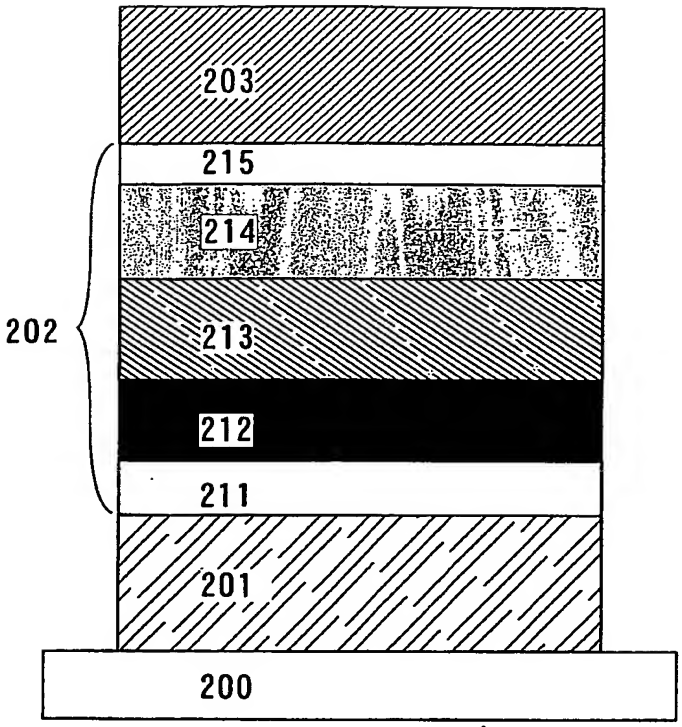


Fig. 14

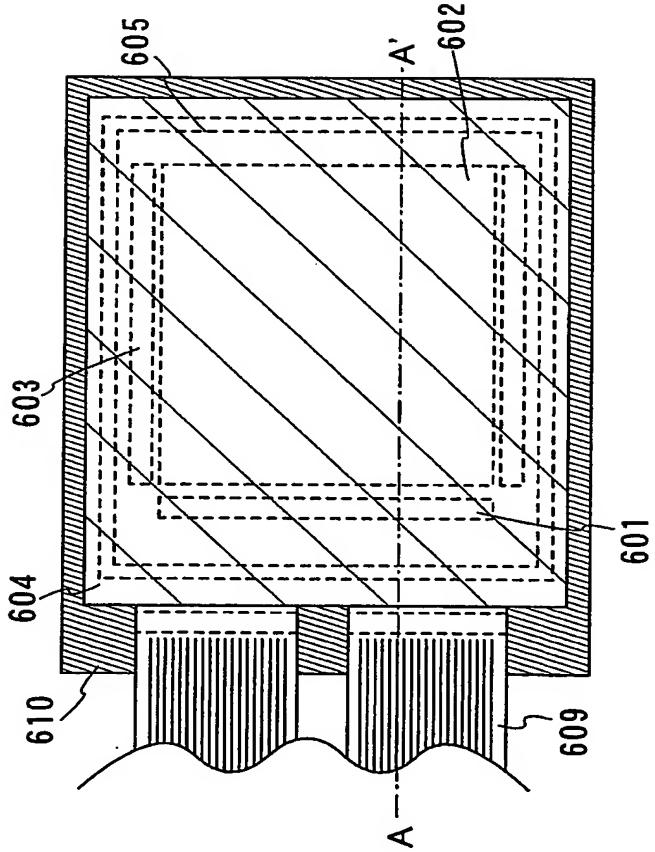


Fig. 15A

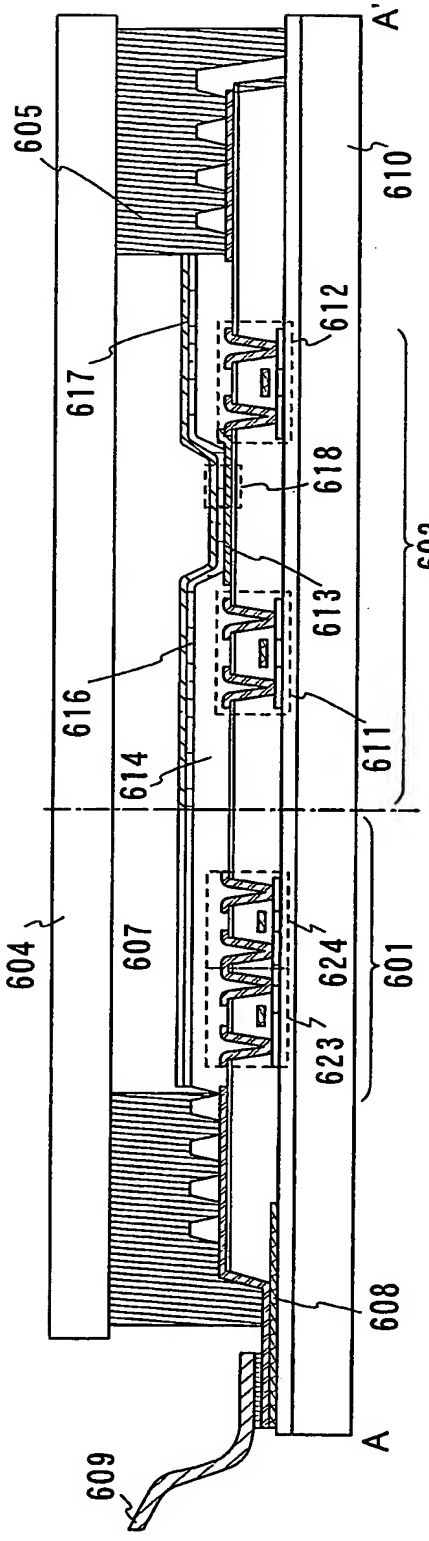


Fig. 15B

Fig. 16A

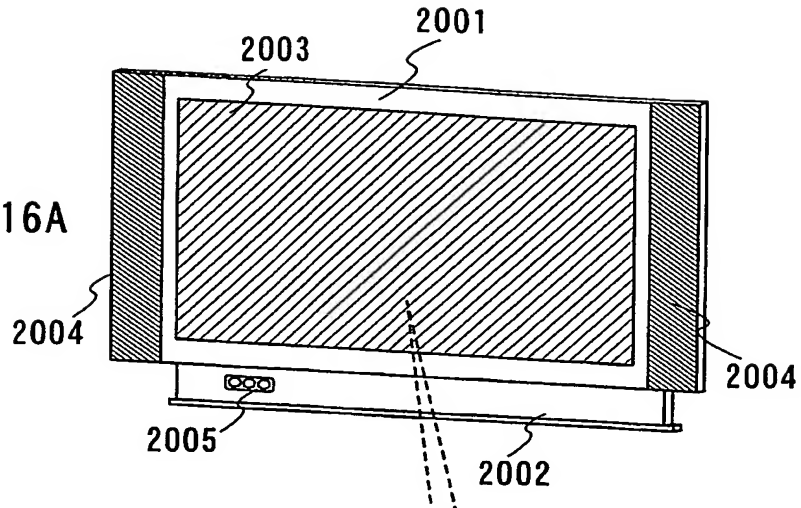


Fig. 16B

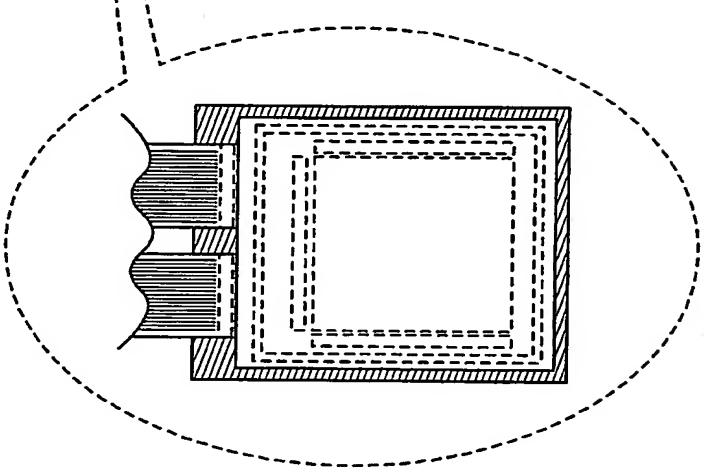
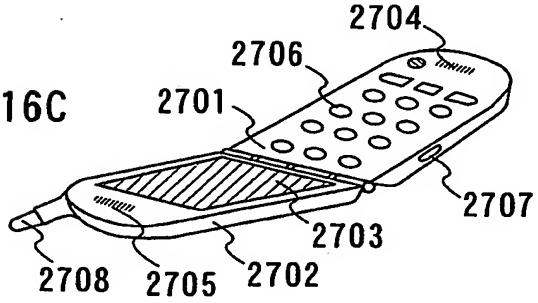


Fig. 16C



EXPLANATION OF SYMBOLS

10: substrate, 11: TFT, 12: TFT, 13: light-emitting element, 14: first electrode, 15: layer including a luminescent material, 16: second electrode, 17: wiring, 100: substrate, 101: first electrode, 102: layer including a luminescent material, 103: second electrode, 111: hole injecting layer, 112: hole transporting layer, 113: light-emitting layer, 114: electron transporting layer, 115: electron injecting layer, 200: substrate, 201: first electrode, 202: layer including a luminescent material, 203: second electrode, 211: hole injecting layer, 212: hole transporting layer, 213: light-emitting layer, 214: electron transporting layer, 215: electron injecting layer, 601: driver circuit portion, 602: pixel portion, 603: driver circuit portion, 604: sealing substrate, 605: sealing material, 607: space, 608: leading wiring, 609: FPC, 610: substrate, 611: switching TFT, 612: current controlling TFT, 613: first electrode, 614: insulator, 616: layer including a luminescent material, 617: second electrode, 618: light-emitting element, 623: n-channel TFT, 624: p-channel TFT, 2001: frame body, 2002: support, 2003: display portion, 2004: speaker portion, 2005: video input terminal, 2701: main body, 2702: frame body, 2703: display portion, 2704: voice input portion, 2705: voice output portion, 2706: operation key, 2707: external connection port, 2708: antenna

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2004/016184

A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl⁷ C07D209/88, C09K11/06, H05B33/22, H05B33/14

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int.Cl⁷ C07D209/88, C09K11/06, H05B33/22, H05B33/14

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
Japanese Utility Model Gazette 1922-1996, Japanese Publication of Unexamined Utility Model Applications 1971-2004, Japanese Registered Utility Model Gazette 1994-2004, Japanese Gazette Containing the Utility Model 1996-2004

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

REGISTRY (STN), CAPLUS (STN), CAOLD (STN)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 2001-279237 A (FUJI PHOTO FILM CO., LTD.) 10.10.2001 see whole document & US 2002/037427 A1	1-8
X	ZHANG, Q. et al., Carbazole-based hole transporting materials for electroluminescent devices, Synthetic Metals, 4 April 2003, Vol.137, p.1111-1112	1-8
X	GRIGALEVICIUS, S. et al., 3,6-Di(N-diphenylamino)-9-phenylcarbazole and its methyl-substituted derivatives as novel hole-transporting amorphous molecular materials, Synthetic Metals, 2002, Vol.128, p.127-131	1

☑ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:
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"E" earlier application or patent but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
07.01.2005	25. 1. 2005

Name and mailing address of the ISA/JP	Authorized officer	
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INTERNATIONALSEARCHREPORT

International application No.
PCT/JP2004/016184

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	GRIGALEVICIUS, S. et al., 3,6-Di(N-diphenylamino)-9-alkylcarbazoles : novel hole-transporting molecular glasses, Synthetic Metals, 2001, Vol.122, p.311-314	1
X	GRIGALEVICIUS, S. et al., Hole-transporting molecular glasses based on carbazole and diphenylamine moieties, Materials Chemistry and Physics, 2001, Vol.72, p.395-400	1
X	JP 2-183259 A (CANON K. K.) 17.07.1990 see whole document (Family:none)	1
X	JP 56-135448 A (XEROX CORP.) 22.10.1981 example IV & JP 59-46249 A & US 4764625 A & CA 1171431 A1	1